ISOTOPES AND RADIATION TECHNOLOGY

A Quarterly Technical Progress Review

Prepared for DIVISION OF TECHNICAL INFORMATION / DIVISION OF ISOTOPES DEVELOPMENT, UNITED STATES ATOMIC ENERGY COMMISSION, by P. S. BAKER, A. F. RUPP, and ASSOCIATES, OAK RIDGE NATIONAL LABORATORY

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TECHNICAL PROGRESS REVIEWS

To meet the needs of industry for concise summaries of current atomic developments, the Atomic Energy Commission is publishing this series, Technical Progress Reviews. Issued quarterly, each of the reviews digests and evaluates the latest findings in a specific area of nuclear technology and science.

The five journals published in this series are:

Isotopes and Radiation Technology, P. S. Baker, A. F. Rupp, and associates, Oak Ridge National Laboratory

Nuclear Safety, Wm. B. Cottrell, W. H. Jordan, and associates, Oak Ridge National Laboratory

Power Reactor Technology, W. H. Zinn and J. R. Dietrich, General Nuclear Engineering Corporation

Reactor Fuel Processing, Stephen Lawroski and associates, Chemical Engineering Division, Argonne National Laboratory

Reactor Materials, R. W. Dayton, E. M. Simons, and associates, Battelle Memorial Institute

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ISOTOPES AND RADIATION TECHNOLOGY

A REVIEW OF RECENT DEVELOPMENTS

Prepared for DIVISION OF TECHNICAL INFORMATION / DIVISION OF ISOTOPES DEVELOPMENT, UNITED STATES ATOMIC ENERGY COMMISSION, by P. S. BAKER, A. F. RUPP, and ASSOCIATES, OAK RIDGE NATIONAL LABORATORY

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"We hear most about atomic armaments and great nuclear reactors that produce large amounts of electricity. But ... atomic energy has a personal, little-known role in your daily life through the extensive and growing application of radioisotopes. ... I am convinced, from the record of the past two decades, that we are only at the beginning of the realization of the power of the atom to increase the prosperity and well-being of people all over the world."

Dr. GLENN T. SEABORG, Chairman
U. S. Atomic Energy Commission
(From "Radioisotopes — A New Servant of Mankind"; paper presented on Oct. 20, 1961, in Tyler, Tex.)

The research and development work sponsored by the federal government generates a vast amount of technical information. There is a pressing need to find effective ways for transmitting this information to those who may have an interest in it. This publication attempts to meet the need for information generated by U. S. Atomic Energy Commission-sponsored research in the field of radioisotopes and radiation technology.

The extent of interest in this field is indicated by the fact that, at the end of 1962, there were about 6000 organizations or individuals in the United States licensed to use radioisotopes.

We hope that this Review will be of service both to present users and to the increasing numbers who will be using radioisotopes in the future.

PAUL C. AEBERSOLD, Director
Division of Isotopes Development
U. S. Atomic Energy Commission

EDWARD J. BRUNENKANT, Director
Division of Technical Information
U. S. Atomic Energy Commission

The recent report of the President's Science Advisory Committee, *Science, Government, and Information*, stresses the importance of the interpretive review journal as a means of keeping our exploding scientific literature from engulfing us. To quote from this report, "Because there are so many technical reports, and most of them are unrefereed and of uneven quality, reviews of technical reports can be particularly useful as discriminating guides to the literature. The reviewer of the report literature must not only tell a coherent story; he must also serve as referee, weeding out what he believes to be wrong from what he believes to be right. Moreover, to make his review fully useful he must cover relevant open literature."

Isotopes and Radiation Technology appears at a time when we at once appreciate fully the size of the information crisis and realize that the critical, interpretive review journal is an essential key to the resolution of the information dilemma. I believe that all those interested in isotopes and radiation technology owe a debt of gratitude to Dr. P. S. Baker and his staff for undertaking the onerous but useful task of making sense of the flood of literature in this field. To the staff of *Isotopes and Radiation Technology* I offer my best wishes for success in this most worthwhile new editorial venture.

ALVIN M. WEINBERG, Director
Oak Ridge National Laboratory

Foreword

This new quarterly, *Isotopes and Radiation Technology*, was approved by the U. S. Atomic Energy Commission (AEC) in the summer of 1962, and its preparation was assigned to Oak Ridge National Laboratory (ORNL) as a part of the general program of the Isotopes Development Center. *Isotopes and Radiation Technology* is one of the series of five AEC-sponsored quarterly Technical Progress Reviews that cover broadly the field of nuclear science and technology. The others in the series are *Nuclear Safety, Power Reactor Technology, Reactor Fuel Processing, and Reactor Materials.*

This Review will emphasize the reporting of Division of Isotopes Development sponsored research and development and will relate this to significant world developments in radioisotopes and radiation technology. By publicizing present applications, it can help to expand the utilization of isotopes; and, by helping the Commission and contractors to keep abreast of related activities within the AEC complex, it can help prevent overlap of research effort and can aid in predicting isotope usage so as to point the direction for future isotope production efforts.

Isotopes and Radiation Technology is a technical progress *review.* Even guest and feature articles will be of a review nature. The editors and the AEC have no intention of duplicating the present efforts of commercial publishers of abstract journals and periodicals covering original research, and overlapping will be avoided.

The ultimate success of the review will, of course, depend upon its value to the readers; a little time and experience will be needed to evaluate this. Therefore we shall make no attempt to fix the format of the quarterly at the present time, and the editors will be counting heavily on reader criticism and suggestions to help them out.

P. S. BAKER, ROBERT H. LAFFERTY, JR., and MARTHA GERRARD, Editors
A. F. RUPP, Advisory Editor
Oak Ridge National Laboratory

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Contents

Foreword

STATUS OF THE U. S. PROGRAM ON ISOTOPES AND RADIATION DEVELOPMENT 1

I INTRODUCTION TO ISOTOPE TECHNOLOGY 27
Isotope Production 28
Isotope Utilization 28
Chronological Outline of Development of Isotopes Program 31
References 34

II ISOTOPE PRODUCTION AND DEVELOPMENT 35
Fission Products 35
Stable Isotopes 43
References 50

III ISOTOPE TECHNOLOGY DEVELOPMENT 53
Analysis 53
References 75

IV PROCESS RADIATION DEVELOPMENT 78
Radiation Engineering Activities at Brookhaven National Laboratory 78
References 82

V SOURCE DEVELOPMENT 84
Glasses 84
References 88

VI ISOTOPIC POWER DEVELOPMENT . . 89
Radioisotope Power Sources 89
Calorimetry for Measuring Radioactivity . . 92
References 95

VII APPLICATIONS IN MEDICINE . . . 97

VIII SAFETY 114

IX AVAILABLE MATERIALS AND SERVICES 122

Issued quarterly by the U. S. Atomic Energy Commission. Use of funds for printing this publication approved by the Director of the Bureau of the Budget on December 31, 1962.

The following special report, which gives an authoritative summary of the status of U. S. Atomic Energy Commission research and development activities, is presented as written, excerpts having been presented at the Fifth Japan Conference on Radioisotopes, Tokyo, Japan, May 21–23, 1963.

Status of the U. S. Program on Isotopes and Radiation Development

By E. E. Fowler and P. C. Aebersold*

Introduction

Major technological advances are being made in the United States to extend and accelerate the beneficial applications of radioisotopes and radiation. The program of the government, carried out by the Atomic Energy Commission (AEC), is contributing importantly to the overall progress in the United States on the development of radioisotopes and radiation technology. The AEC program is directed toward (1) advancing radioisotopes and radiation technology to encourage development of beneficial applications, particularly those to meet problems of urgent public interest; (2) accelerating realization of the potential contribution of radioisotopes and radiation applications to the national economy and welfare; and (3) providing an important contribution to world development in the peaceful uses of atomic energy.

These objectives are being achieved through a multiphase research and development program including the following major elements:

1. Radioisotope and Fission-Product Production and Separations Technology fulfills the AEC's responsibility for ensuring the availability of radioisotopes and radiation sources to meet the changing needs of advancing science and technology in the United States.

2. Isotopic Power Fuels Development is directed to the development and production of isotopic fuels and fuel forms for a broad spectrum of thermal applications, including the SNAP auxiliary-electric-power devices.

3. Process Radiation Development fosters development of a broad technology leading to productive use of megacurie quantities of fission products and other radioisotopes for process radiation purposes.

4. Radiation Pasteurization of Foods involves the development of technology for low-dose radiation processing of certain fruit and fishery products to extend their refrigerated shelf life.

5. Radioisotope Technology Development creates a broad base of new and improved technology required for extending and speeding up the application of radioisotopes in science and technology.

The progress and scope of activities in each of these areas over the last year have been impressive.

In the production of radioisotopes, advances continue to be made through development of processes for producing new isotopes and chemical forms, as well as high radiochemically pure materials, to meet the needs of science and technology in the United States. We are now routinely producing multimegacurie quantities of Co^{60}, Sr^{90}, and Cs^{137} for process radiation-application developments and for use as thermal sources for devices to power automatic weather stations, light buoys, seismographic units, and underseas electronic equipment. During the past year the world's largest single Co^{60} source, approximately 1.3 million curies, was produced

*Division of Isotopes Development, United States Atomic Energy Commission, Washington, D. C.

at the AEC's Savannah River production site and delivered to the U. S. Army Radiation Laboratory at Natick, Mass. Included in our current large-source production effort are 120,000 curies of Co^{60} for the Japan Atomic Energy Research Institute. The requirement for reliable, long-term auxiliary electric power in the national space program has resulted in a stepped-up development effort in the production of large quantities of previously scarce radioisotopes such as Pu^{238}, Cm^{242}, and Cm^{244}. These radioisotope power sources are planned for use in lunar scientific probes and in communications and meteorological satellites.

In the United States an estimated 50 kw of installed radiation power is now being used for commercial process radiation purposes. The value of production is estimated at $20 million annually. We have witnessed the emergence on the commercial market of new radiation-produced products which can either be produced better by radiation or which cannot be made readily by other means. The first industrial use of radiation for bulk chemical manufacture in the United States is now being carried out by the Dow Chemical Company. This involves the commercial-scale production of ethyl bromide, using Co^{60} as the source of radiation energy. Significant advances made in the Commission's process radiation development program range from radiation polymerization of wood impregnated with monomer materials to form "wood-plastic alloys" to the discovery of a new reaction mechanism for formation of hydrazine by irradiation of gaseous ammonia.

Significant achievements have been made in developing the technology for radiation pasteurization of foods, including processing conditions needed to ensure the retention of freshness and nutritional values. In addition, preliminary marketing and economic studies are being made in anticipation of the ultimate commercialization of the process.

In the area of radioisotope technology, advances range from developments leading to an ocean-bottom-sediment density meter to an activation-analysis technique for analyzing the surface of the moon. A significant new development is a portable beta-excited X-ray system which uses a Pm^{147} radioisotope source and film-developing unit so compact that both can be held in one hand. Neutron activation analytic techniques have been greatly expanded in range of application from identification of manufactured materials, such as plastics, to scientific crime detection.

The AEC's research and development work on isotopes and radiation is managed by its Division of Isotopes Development and carried out in the AEC's national laboratories and with private research laboratories. Radioisotopes and radiation research and development projects now being carried out for the AEC by private research groups are listed below.

LIST OF RESEARCH AND DEVELOPMENT CONTRACTS,
DIVISION OF ISOTOPES DEVELOPMENT,
UNITED STATES ATOMIC ENERGY COMMISSION

Process Radiation Development

AIR REDUCTION COMPANY, INC. Murray Hill, N. J.	Investigation of the initiation of chemical reactions using Kr^{85} beta radiation
ARMOUR RESEARCH FOUNDATION Chicago, Ill. (Now called Illinois Institute of Technology Research Institute.)	Study of radiation reaction mechanism producing high-value halogenated aromatic compounds
	Study of ultrahigh-intensity radiation effects
ATLANTIC RESEARCH CORP. Alexandria, Va.	Study of the use of organometallic bond in radiochemical dosimetry
BATTELLE MEMORIAL INSTITUTE Columbus, Ohio	Study of structural and compositional factors influencing radiation-induced polymerization reactions
CONSOLIDATION COAL CO. Pittsburgh, Pa.	Evaluation of beta radiation as a hydrogenation catalyst
FUNDAMENTAL METHODS ASSOC. New York, N. Y.	Research on neutron transmutation doping

GEORGIA TECHNOLOGICAL RESEARCH INSTITUTE Atlanta, Ga.	Effects of high-intensity ionizing radiation on colloidal systems and suspensions
	Physical properties and structural characteristics of polymers resulting from post-effect polymerization
RADIATION APPLICATIONS INC. Long Island City, N. Y.	Study of the mechanism of radiation-induced gelation in monomer-polymer mixtures
STANFORD RESEARCH INSTITUTE Menlo Park, Calif.	Radiation-induced reactions in swollen polymers
TEXTILE RESEARCH CENTER Raleigh, N. C.	Applications of nuclear radiation and radioisotope techniques to textile materials and processes
WESTINGHOUSE ELECTRIC CORP. Pittsburgh, Pa.	Investigation of stimulated emission of gamma radiation
WEST VIRGINIA UNIVERSITY Morgantown, W. Va.	Development of wood-plastic combinations using gamma radiation to induce polymerization
WILLIAM H. JOHNSTON LABS., INC., Baltimore, Md.	Mass spectrometry of fast primary ion reactions

Radiation Pasteurization of Foods

AGRICULTURAL MARKETING SERVICE U. S. Department of Agriculture, Washington, D. C.	Studies on gamma-radiation treatments for the control of postharvest diseases of fresh fruits and vegetables
BUREAU OF COMMERCIAL FISHERIES U. S. Department of the Interior, Gloucester, Mass.	Research to conduct quality studies on several east-coast seafoods, to include determination of process parameters, flavor and odor studies, effect of preirradiation quality on postirradiation storage, and development of appropriate packaging containers
	Research on the radiation pasteurization of Dungeness crab meat and flounder
ECONOMIC RESEARCH SERVICE U. S. Department of Agriculture, Washington, D. C.	Economic feasibility of radiation pasteurization of selected fruits and vegetables
HAZELTON LABORATORIES Falls Church, Va.	Extractive studies on packaging material to be used with irradiated foods
LOUISIANA STATE UNIVERSITY Baton Rouge, La.	Radiation pasteurization of shrimp
MASSACHUSETTS INSTITUTE OF TECHNOLOGY Department of Food Technology, Cambridge, Mass.	Study of the effects of radiation pasteurization on the storage-life extension of soft-shelled clams and haddock fillets
UNIVERSITY OF CALIFORNIA Department of Pomology, Davis, Calif.	Research on radiation pasteurization of fruits and vegetables
UNIVERSITY OF WASHINGTON Seattle 5, Wash.	Study of radiation pasteurization of marine products and installation and operation of Co^{60} gamma research irradiator

Isotopes Technology Development

ARMOUR RESEARCH FOUNDATION Chicago, Ill. (Now called Illinois Institute of Technology Research Institute.)	Scintillation spectrometer measurements of capture gamma rays from natural elements

BATTELLE MEMORIAL INSTITUTE Columbus, Ohio	Development and evaluation of safety performance criteria for sealed radiation sources
BUREAU OF MINES U. S. Department of the Interior, Washington 25, D. C.	Research on radioactive tracers to evaluate heterogeneities in repressured petroleum reservoirs
CATHOLIC UNIVERSITY OF AMERICA Washington 17, D. C.	Investigations regarding limiting factors in rapid identification of selected materials by nuclear techniques
CLEGG RESEARCH LABORATORIES Santa Fe, N. Mex.	Absorptiometry with beta-excited X rays
COLORADO SCHOOL OF MINES RESEARCH FOUNDATION, INC. Golden, Colo.	Evaluation of large-volume detectors and their application to isotope process control
DRESSER RESEARCH Tulsa, Okla.	Development of an ocean-bottom-sediment density meter and a shipboard neutron activation analyzer for ocean-bottom samples
EDSEL B. FORD INSTITUTE Detroit, Mich.	Compilation of beta-excited X-ray source spectra
FOREST SERVICE U. S. Department of Agriculture, Berkeley, Calif.	Use of radioactive isotopes in measurement of snow density profiles
GENERAL ATOMIC Division of General Dynamics Corp., San Diego, Calif.	Neutron activation analysis in scientific crime detection
	Development of techniques for utilizing reactor fast neutrons for activation analysis
GEOLOGICAL SURVEY U. S. Department of the Interior, Washington 25, D. C.	Development of radioisotope techniques for water-resource studies
INTERNAL REVENUE SERVICE Alcohol & Tobacco Tax Div., U. S. Treasury Department, Washington 25, D. C.	Evaluation of the effectiveness of neutron radioactivation analysis in criminalistics
MARTIN-MARIETTA CORP. Baltimore, Md.	Development of radioisotope-energized thermally actuated underwater acoustic device
MB ASSOCIATES Walnut Creek, Calif.	Alpha detector
MULLER-MONROE DESIGNS Santa Fe, N. Mex.	Investigation of methods for producing and measuring beta-excited X-ray diffraction effects
NATIONAL CANNERS ASSOCIATION Berkeley 10, Calif.	Investigations of radioisotope tracer techniques to detect and measure detergent residues on washed food products
	Development of radioisotope techniques to evaluate and improve routine practices for removal of pesticides from food
PARAMETRICS, INC. Saxonville, Mass.	Development of Kr^{85} as a universal tracer
	Feasibility study of radiological mechanisms for use in geophysics research
RESEARCH TRIANGLE INSTITUTE Durham, N. C.	Low-level tracer studies, fission-product uses, soil-moisture studies, and quality-control techniques

TEXAS NUCLEAR CORPORATION Austin, Tex.	Investigation of the feasibility of using nuclear techniques for coal analysis
TRACERLAB INC. Waltham, Mass.	Radioisotope application to printed-circuit board technology
	Development of narrow-band beta-excited X-ray detector for analysis of chemical elements in complex matrices
UNDERWRITERS' LABORATORIES, INC. Chicago, Ill.	Fire-resistance tests of teletherapy unit and shipping shields
UNION CARBIDE NUCLEAR CO. Research Center, Tuxedo, N. Y.	Investigation of radiochemical separations and metastable isomers for use in activation analysis
UNIVERSITY OF CHICAGO Chicago, Ill.	Radioisotope research, development, and related activities designed to lead to new or improved techniques or applications of radioisotopes to scientific, industrial, and public-benefit problems
WESTINGHOUSE ELECTRIC CORP. Pittsburgh, Pa.	Study of applications of the Mossbauer effects to explore its potential to public benefit
WILLIAM H. JOHNSTON LABS., INC. Baltimore 15, Md.	Development and testing of a deep-water isotopic current analyzer
Production and Materials	
MARTIN-MARIETTA CORP. Baltimore, Md.	Isotopic power fuels research and development

Radioisotope and Fission-Product Production and Separations Technology

In certain respects, the AEC's work on radioisotopes and fission-product production and materials development is probably one of its most important activities. This is based on the fact that all its radioisotopes and radiation technology development effort must ultimately be based on a continuing availability of suitable radioactive materials in adequate quantities and at a reasonable price.

The changing needs of science and technology have presented a continuing challenge to our radioisotopes production program. Today 137 individual radioisotopes are routinely available from the AEC. On almost a day-to-day basis, efforts are directed toward development of new radioisotopes and radiation sources, as well as toward improved production processes for existing radioisotopes. Improved production is particularly important because many recent industrial and medical applications require ever-increasing quantities of radioactive materials. For example, as of Jan. 1, 1963, Oak Ridge National Laboratory had produced and distributed about 2.1 million curies of radioactive materials during the entire 16 years of the radioisotopes distribution program. However, almost one-half of this amount, or about one million curies, was distributed during just the past two years. This substantial growth in the demand for radioactive materials and associated services and equipment has also led to the development of a substantial private radioisotopes business in the United States. The market for modified isotope products and devices in 1962 was in the $27 million to $34 million range. The following is a breakdown according to product areas:

- —current sales of labeled organic compounds are $3 million to $4 million per year. Growth rate is at a level of approximately 10 to 15% per year.
- —current sales for radiopharmaceuticals are at a $5 million level. Growth rate is at a level of approximately 25% per year.
- —sales of teletherapy units have reached the $2 million to $3 million per year level.

Seven U. S. manufacturers produce these devices.

—the market for radiographic equipment and commercial irradiation services has reached the $15 million to $20 million per year level.

Several developments in the area of new radioisotopes production should be mentioned. One of the most promising is the use of enriched targets in radioisotope production. For example, Hf^{175}, a low-energy gamma emitter with a 70-day half-life was produced by the reactor irradiation of a Hf^{174} target enriched to 7.85% Hf^{174}. Similarly, Ge^{71} was produced through the irradiation of 86% enriched Ge^{70}. Tin-119*m* was made by the irradiation of a target enriched in Sn^{118}.

Another area is the development of X-ray and bremsstrahlung radiation sources, tailor-made to emit a specific energy X ray for a specific purpose. We have prepared Pm^{147} pellets encapsulated in aluminum to form sources of low-energy X rays. Beta emission from the decay of Pm^{147} produces its characteristic 38-kev *K* X ray as secondary radiation. In the same manner, the characteristic 30-kev X ray of antimony can be produced by pressing a mixture of antimony metal and promethium oxide into a pellet. Promethium-147 bremsstrahlung sources have also been fabricated with platinum sponge and tungsten carbide. Three hundred millicuries of Pm^{147} adsorbed on Decalso and mixed with 1 g of platinum gave 1.7×10^8 photons per minute of the characteristic 67-kev platinum X ray. The same quantity of Pm^{147} adsorbed on Decalso and mixed with 2 g of tungsten carbide emits 1.7×10^8 photons per minute of the characteristic 59-kev tungsten X ray.

Substantial progress is also being made in techniques for the preparation of isotopically enriched materials. For example, we expect soon to be prepared to offer Kr^{85} enriched up to as much as 50%.

Another potentially valuable development we are actively following is the use of I^{125} sources for medical and industrial X-ray radiography.

During the past year Oak Ridge National Laboratory (ORNL) has been intensively studying the preparation and encapsulation of I^{125} X-ray sources. At first the compound used was silver iodide. The source was prepared by precipitating silver iodide with the minimum iodide carrier added and then centrifuging and drying the precipitate. The precipitate containing 2 curies of I^{125} was encapsulated in stainless steel having a 2.5-mm inner diameter and a 1-mil-thick window. Evaluation of the use of I^{125} as a medical radiography source has been made at the Ohio State University.

It was determined, as a result of the evaluation, that the efficiency of the emission was decreased because of the high *K* absorption edge of silver for the characteristic I^{125} gamma photon. To improve the output, sodium iodide was used as the compound for a second source, which is now being evaluated.

Oak Ridge National Laboratory has also investigated plating I^{125} on platinum, silver, and palladium by chemical reaction. Only limited deposition of iodine was obtained. Also, the deposited activity rubbed off easily and could be released upon heating. We are therefore presently examining the adsorption of I^{125} vapor on a tiny porous carbon plug and encapsulating it in stainless steel with an electroplated nickel end window 1 mil thick.

Proceeding apace with I^{125} source fabrication is the development of improved production techniques to reduce cost. Oak Ridge National Laboratory is now preparing to produce enriched Xe^{124} target materials in order to increase yields and is examining the installation of a loop in the Oak Ridge Research Reactor for continuous production of I^{125}.

Isotopic Power Fuels Development

A most exciting new area of radioisotopes application is based on the liberation of thermal energy during radioisotope decay. This has led to the development of small nuclear power supplies, which derive their energy from radioisotope decay, for use in locations and under environments where more conventional sources of electric power are unsatisfactory.

Isotopic power systems, or SNAP devices as they are called, are now being used to power navigational satellites in space, remote automatic weather stations near both the North and South Poles, and navigational buoys and underseas electronic equipment. Other SNAP devices will probably soon power barge-mounted automatic weather stations, lunar scientific probes, communications and meteorological satellites,

and a host of additional terrestrial, marine, and space systems.

Present indications are that by 1970 many electrical kilowatts of radioisotope power will be required annually. Consequently the AEC program has as its ultimate objective the yearly production of perhaps as much as 1000 kw(t) of radioisotope materials. Tables 1 and 2 identify present and planned production goals.

The incredibly large quantities of radioactive materials, possessing appropriate characteristics, which will be required to satisfy isotopic power needs have posed the most severe challenge to the AEC's materials development and production activity since its inception in 1946.

Two major categories of radioisotopes are being considered as heat sources: the beta-emitting fission products separated from gross radioactive wastes generated in the nuclear fission process, and certain very energetic alpha emitters.

It is important to note that several hundred individually identifiable isotopes are produced by nuclear fission, but only a relatively few have characteristics suitable for thermal application. These characteristics are:

1. A reasonably long half-life
2. A high-enough yield in the fission process to make substantial quantities available at reasonable cost
3. A capability of being formed into high-power-density heat sources
4. A decay that ideally does not involve the emission of high-energy gamma rays

In the familiar double-humped yield curve for the thermal fission of U^{235}, the peak occurs at mass numbers of roughly 90 and 140, and isotopes in yields of around 5% will be found in the vicinity of these numbers. Thus Sr^{90}, Cs^{137}, Ce^{144}, and Pm^{147} are the only radioactive fission products with half-lives greater than six months and less than 100 years that are of interest to us.

The potential alpha emitters, which are produced by specific reactor irradiation, must be selected on the basis of the same general criteria (except fission yield) as the fission products. Only Po^{210}, Pu^{238}, Cm^{242}, and Cm^{244} will meet these criteria. The quantities of these materials which can be produced are, of course, limited only by the availability of target materials, reactor space, and chemical processing facilities.

In almost every instance the elemental forms of the radioisotopes are not considered the most desirable for heat sources. Rather they are made into compounds exhibiting physical and chemical properties required for particular application. Ordinarily it is desirable to concentrate as much radioactivity as possible in a given volume. Thus an important criterion in evaluating the usefulness of an isotope compound will be its "power density" expressed in units of watts per cubic centimeter. This unit in turn is the product of the specific power in watts per gram and the material density in grams per cubic centimeter.

The radioisotopes of interest are summarized in Table 3.

The chemical form of the radioisotope must possess other desirable properties if it is to be useful for thermal-source applications. Certainly its melting point should be higher than the maximum temperature it will experience in an operational situation. It should be chemically inert with respect to the materials used to encapsulate it. It should be stable at the expected operating temperatures and not decompose or swell in an unusual fashion. Its thermal conductivity should be high enough to permit escape of the heat generated without undue temperature differentials through the compound. It should be relatively insoluble in

Table 1 RADIOISOTOPE PRODUCTION CAPABILITY FOR THERMAL APPLICATIONS

Isotope	1963	1964	1965	1966	1967	1968	1969	1970	1971	1972
Sr^{90}, megacuries	3	5	5	5	10	10	10	10	10	10
Cs^{137}, megacuries	1	3.5	3.5	3.5	10	10	10	10	10	10
Ce^{144}, megacuries	3.5	3.5	3.5	3.5	100	100	100	100	100	100
Pm^{147}, megacuries	0.02	0.5	0.5	0.5	30	30	30	30	30	30
Pu^{238}, kg	3	6	13	18	24	32	36	42	47	51
Cm^{244}, kg		0.05	0.1	6	18	40	56	56	58	58
Cm^{242}, g		12	80	1000	1000	1000	1000	1000	1000	1000
Po^{210}, g	20	50	100	100	1000	1000	1000	1000	1000	1000

naturally occurring fluids, such as sea water and fresh water, if it is to be employed for terrestrial and marine applications; and it should be of a form that is readily dispersible upon reentry heating if it is employed in space missions. Last, it must be a compound or mixture that is readily fabricable by remote techniques in a hot cell.

This discussion of criteria used in the selection of appropriate radioactive materials for thermal applications provides some idea of the constraints placed upon the AEC both in materials development and in production.

Today the AEC's Hanford Atomic Products Operation is the only source for fission products in the United States. Until reprocessing plants are built specifically for handling power-reactor fuel elements, Hanford will probably remain the only source. Several years ago, when the need for large quantities of separated fission products was recognized, interim facilities for the separation of the four principal fission products (Sr^{90}, Cs^{137}, Ce^{144}, and Pm^{147}) were installed at Hanford. These crude cuts are then shipped to ORNL and the Martin Company facility at Quehanna, Pa., for further purification, processing, and encapsulation. This interim arrangement, nevertheless, gives us an annual capability for producing multimegacurie quantities of final product. At Hanford a new concept of radioactive waste management has been proposed. It would ensure the continuing availability of greatly increased quantities of the four principal fission products. These isotopes would be removed from the gross fission-product stream, and the remaining wastes would be solidified in storage tanks by evaporation. The separated long-lived fission products could then either be purified and processed for beneficial utilization or adsorbed on an inorganic ion-exchange material and sealed in stainless-steel cans for permanent storage. Such a plan would alleviate present difficulties resulting from self-heating wastes in storage caused by the long-lived fission products.

Hanford also now is studying the design of a fully integrated fission-products processing, fabrication, and encapsulating facility. Such a facility could convert and package for utilization all the Sr^{90}, Cs^{137}, Ce^{144}, and Pm^{147} generated in the operation of the Hanford reactors. The design study is not complete, but preliminary data indicate that we can make available, annually, well over one hundred million curies of packaged fission products at only a fraction of today's costs.

In addition to efforts in improving and optimizing fission-products production, significant emphasis is also being placed on the development of improved materials. For example, although strontium titanate is an excellent material for terrestrial and marine applications, it does not have the appropriate characteristics for space applications. Research and development work is under way on the preparation of a Sr^{90} fuel form for future space use. The program is expected to yield a form of strontium that will not only meet the rigid safety requirements set for space nuclear systems but will possess a significantly higher power density than strontium titanate. This latter characteristic is an absolute one for those applications where maximum power-to-weight ratio is essential.

Efforts are continuing apace in the development of improved fuel forms for Cs^{137} and Ce^{144}.

Table 2 THERMAL POWER AVAILABILITY FROM RADIOISOTOPES*

(Kilowatts per Year at Fuel Fabrication Time)

Isotope	1963	1964	1965	1966	1967	1968	1969	1970	1971	1972
Sr^{90}	19	32	32	32	63	63	63	63	63	63
Cs^{137}	5	17	17	17	48	48	48	48	48	48
Ce^{144}	25	25	25	25	700	700	700	700	700	700
Pm^{147}	0.01	0.2	0.2	0.2	14	14	14	14	14	14
Pu^{238}	1.5	3.0	6.0	0.5	11.5	15	17	20	23	25
Cm^{244}		0.1	0.2	14	41	92	129	129	134	134
Cm^{242}		1.5	9.5	120	120	120	120	120	120	120
Po^{210}	3.0	7.0	14	14	140	140	140	140	140	140

*Based on quantities of material in Table 1.

Table 3 PROPERTIES OF RADIOISOTOPES FOR ISOTOPIC POWER APPLICATIONS

Isotope	Type of decay	Half-life	Chemical form	Specific thermal power: Watts/kilocurie	Specific thermal power: Watts/g	Specific thermal power: Watts/cm³	Material requirement for mission: Mission life, years	Material requirement for mission: Curies/electrical watt
Sr^{90}	Beta	28 years	Titanate	6.5	0.2	0.7	10	4,000
Cs^{137}	Beta Gamma	27 years	Glass	4.8	0.072	0.22	10	5,500
Ce^{144}	Beta Gamma	285 days	Oxide	7.9	2.3	13.8	1	7,000
Pm^{147}	Beta	2.6 years	Oxide	0.37	0.18	1.0	2.6	108,000
Pu^{238}	Alpha	89.6 years	Metal	34.5	0.48	9.3	10	625
Cm^{244}	Alpha	18.4 years	Oxide	35.0	2.3	22.4	10	840
Cm^{242}	Alpha	163 days	Oxide	36.2	120	1170	0.5	1,190
Po^{210}	Alpha	138 days	Metal	31.7	140	1320	0.5	1,550

The presently accepted product for Ce^{144}, ceric oxide, while possessing very high power densities and temperature stability, liberates oxygen on radioactive decay. This poses problems in the design of appropriate encapsulated thermal sources. We have, therefore, a continuing program aimed at the development of appropriate oxygen-free forms of Ce^{144}.

A particularly challenging effort is the development of technology leading to the production of large quantities of Cm^{242}. Produced now in only milligram to gram quantities for research purposes, Cm^{242} has been selected as the isotopic fuel for auxiliary power in the Surveyor spacecraft because of its very high power density and easily shielded radiations. The Surveyor mission, designed to land instruments on the moon's surface in support of the Apollo program, will require tens of grams of the Cm^{242} fuel annually. Oak Ridge National Laboratory, responsible for the effort, is now completing the installation of facilities for the production of Am^{241} targets, the solvent-extraction separation of Am^{241}-Cm^{242} fraction from the fission products, and the fabrication of thermal sources. Americium-241 will be pelletized, encapsulated, and irradiated in a high-flux reactor such as the Oak Ridge Research Reactor or the Materials Testing Reactor at the National Reactor Testing Station in Idaho. The irradiated material will be dissolved and the fission products will be removed by solvent extraction, but the Am^{241} will probably not be separated from the Cm^{242}. The final solution will be further processed into a Cm^{242} oxide, which will be calcined, pelletized, sintered, doubly encapsulated, welded, and leak checked. The first substantial quantities of material will probably be produced this year, with our production capability rising by 1964 to at least 80 g annually.

The technology being developed under the Cm^{242} program will be useful in the production of Cm^{244}, a radioisotope that is particularly interesting as an auxiliary-electric-power source for aerospace applications. Again, large-scale production represents a truly pioneering effort. The objectives of the Cm^{244} production program are (1) the development of chemical processes for recovering and purifying a curium-americium mixture from irradiated plutonium targets, (2) the development of the optimum Cm^{244} source compound or mixture for specific objectives, and (3) the determination of the characteristics of Cm^{244} thermal sources. Although chemically identical with Cm^{242}, Cm^{244} processing and utilization present unique problems because of differences in feed-material composition and specific power generation. Applications of Cm^{244} may require its separation from americium, which is not necessary in the case of Cm^{242}, in order to achieve the required power density. Source compounds and mixtures of Cm^{244} will have different requirements from those of Cm^{242} for thermal conductivity, radiation emissions, size, shape, and encapsulation.

Both Po^{210} and Pu^{238} have been routinely produced and utilized as the metal for thermal applications. However, the trend in SNAP application is toward higher efficiency direct-energy conversion systems requiring isotope sources capable of operating at greatly elevated temperatures. Consequently our research has been directed toward developing high-temperature–low-vapor-pressure compounds of polo-

nium and plutonium and toward welding techniques and materials of encapsulation for high-temperature containment. Although our results are not yet conclusive, the family of rare earth–polonium compounds looks most promising, as do some of the ceramic forms of Pu^{238}.

Safety Testing

The remaining major program activity associated with the AEC's radioisotope production efforts relates to safety testing of sealed radioisotope sources. The program will provide a fund of information that can be used by source manufacturers, users, and regulatory bodies to help maintain the present high standards of safety in radioisotope applications.

The first objective in the program is to develop a uniform source-classification system based on the mechanical and environmental integrity of the source. A uniform system will give U. S. regulatory groups consistent methods for defining source characteristics. The classes will be defined wherever possible in terms of numerical values (i.e., impact strength, tensile strength, corrosion rate, etc.) to minimize misinterpretation.

When strength and corrosion values for the various classes of sources are established, standard tests will be developed which can be used to determine the class in which a source belongs. At the same time, simple quality-control tests will be developed which can be used by manufacturers to measure the quality of their product against the requirements of the source class.

The third objective of the program is to develop a rating system that can be used by both manufacturers and regulatory groups to determine the classes of sources required for particular applications. The system will be based on data obtained by testing a variety of sources now in use which have demonstrated their reliability from a safety standpoint, on experience gained in developing classification and quality-control tests, and on results of a survey of environmental conditions existing in various types of source applications.

The survey of environmental conditions has recently been completed by Battelle Memorial Institute under contract to the Atomic Energy Commission. The source-classification system should be available by the end of this year.

In the interim, detailed testing of commercially available sources is proceeding at ORNL. Work is expected to start in 1964 on standardizing quality-control tests along lines compatible with the classification system and tests. By 1965 sufficient data should be available to begin work on the development of a sealed-source rating system.

Another aspect of our safety program is the test evaluation of radioisotope shipping casks and medical teletherapy units to ensure that they adequately meet existing and planned safety criteria. Many drop tests have been conducted during the past year to test the structural integrity of a broad range of shipping casks. The casks are dropped on a pad that consists of a 12- by 12-ft piece of steel armor plate 4.5 in. thick, backed up by a 5-ft-thick slab of reinforced concrete. Below this pad is a 3-ft-diameter reinforced column reaching down 10 ft into bedrock.

Each cask is instrumented with accelerometers, strain gages, and compressometers to measure inner wall deflections. Some of the casks are covered with brittle lacquer material to determine the areas of high stress.

Casks have been dropped horizontally, on end, on a corner, and on pistons. Results indicate the casks will meet free-fall safety requirements, although there is some doubt whether the outer shells of casks weighing more than 6 tons can resist penetration by a 6-in.-diameter piston.

Similarly, we are fire-testing both shipping casks and shielding devices such as medical teletherapy heads to determine if they meet the standard 1-hr fire requirement as established by the National Fire Protection Association.

Process Radiation Development

Contrasted to the United States' slow progress during the past decade toward industrial utilization of radiation for manufacture of chemicals and chemical products, we are today witnessing new gains, new interests, and new effort in developing ionizing radiation for process purposes.

An estimated 50 kw of installed radiation power is now being used for commercial radiation purposes. We are witnessing the emergence on the commercial market of new radiation-

produced products. The first industrial use of radiation for bulk chemical manufacture in the United States is now being carried out by the Dow Chemical Company. This involves the commercial-scale production of ethyl bromide, using Co^{60} as the source of radiation energy. Other products include radiation-crosslinked polyethylene wrap and insulation, radiation-induced graft polymers, radiation-cured industrial coating, radiation-modified semiconductors, and radiation-sterilized medical supplies. Estimates place the commercial process radiation business at $20 million per year. Tables 4 and 5 list the radiation-processed products now being manufactured in the United States or planned for production.

In the United States today, the government, through the Process Radiation Development Program of the AEC, is attempting to provide a productive, balanced effort on radiation processing in concert with that of private industry. The AEC's Process Radiation Development Program is designed and directed toward:

1. Seeking original radiation reactions, thereby establishing a substantial scientific foundation leading ultimately to production of unique radiation products
2. Establishing the technical feasibility of specific process radiation systems
3. Learning to design, construct, and operate large kilowatt radiation processing facilities
4. Finding ways of cutting radiation costs
5. Developing large-scale requirements for fission products and other radioisotopes in the process radiation field

The major areas of program work include:

1. Radiation Engineering — studies devoted to problems of radiation source and facility design, fabrication, and construction necessary for the support of general and specific radiation processes

2. Exploratory Processing Systems — studies conducted on an exploratory or "scouting" basis with the most promising systems being developed to the point of demonstrating technical feasibility

3. Advanced Development Systems — engineering scale-up projects designed to establish economic feasibility of those processes which show the greatest and most immediate commercial promise

4. Dosimetry — studies necessary to provide dosimetric systems for eventual use in a radiation process. These systems can be considered primarily in terms of quality control.

During the past year the AEC has completed and placed into operational use its Radiation Development Center at BNL. This new laboratory is one of the world's most versatile and unique radiation facilities. It will serve as the center for government work on process radiation engineering, the science of handling and using large radiation sources for processing purposes. The laboratory is a unique canal-cell complex. It consists of a preparation cell for

Table 4 EXISTING COMMERCIAL RADIATION PROCESSES

Process	Source	Company	Volume
Production of ethyl bromide	Co^{60}	Dow Chemical Co.	400 tons per year
Polyethylene film	Machine	W. R. Grace & Co.	1500 tons per year
Crosslinked polyethylene wire		Radiation Applications Inc.	
Insulation, circuit boards, etc.	Machine	General Electric Co. Raychem (Redwood City, Calif.)	$15 million per year
Medical-supply sterilization	Machine Co^{60}	Ethicon, Inc. Hospital Supply Co., Inc. Becton-Dickinson & Co.	80% of USA market (sutures)
Goat-hair sterilization	Co^{60}	Westminster Carpet Co. (Australia)	2250 tons per year
Sprout inhibition on potatoes	Co^{60}	Atomic Energy of Canada, Ltd.	500 tons per year
Production of polyvinyl acetal resin	Machine	Toyo Rayon (Japan)	No estimate
Semiconductors	Machine	At least six of the larger semiconductor producers	15 to 20 million units per year

building large radiation sources into desired configurations and an experimental cell that will permit mockup of process systems to investigate such problems as source geometry, radiation dose distribution, and process radiation utilization efficiencies. The Radiation Development Center will be capable of handling several million curies of Co^{60} or its equivalent. By summer 1963 it is expected that 600 kilocuries of Co^{60} will be assembled and installed in the Center for use. We plan to increase the size of the Co^{60} source up to 3 million curies. We are also completing fabrication of a 200-kilocurie Cs^{137} source, which will be installed in the Radiation Development Center. As work progresses, other radioisotopes, fission products, and machine radiation sources will be studied and compared.

Research in the Radiation Development Center is intended to provide data leading to a radiation engineering handbook. This handbook, to be published in its first-edition form by 1964, will contain information on basic radiation physics, source design and fabrication, computational procedures, safety aspects, health physics, shielding compilations, and economic guidelines. Our purpose is to encourage use of the Radiation Development Center to provide experience in radiation engineering problems, on a research participation basis, to government and industry scientists as well as scientists from other countries. The Radiation Development Center can thus serve as the major focal point for radiation development in the United States.

Within the scope of this report, it is not practical to provide a comprehensive discussion of research and development being conducted under the AEC's Process Radiation Development Program.* Therefore, with the exception of brief identification of overall process work, discussion will be limited to those process radiation systems which now appear most promising for ultimate development.

Table 5 NEAR-FUTURE COMMERCIAL RADIATION PROCESSES

Process	Probable source	Potential use
Radiation curing of coatings	Machine	Plastic-coated plywood panels and paint-primer curing
Textiles	Machine	Improvement of dyeability, increased mildew and water resistance, fire retardant
Wood-plastic alloys	Machine Co^{60}	Wood construction, furniture, piling, and sporting equipment
Sterilization of medical supplies	Co^{60}	Bulk-packaged sutures, catheter kits, etc.
Food pasteurization	Co^{60}	Extension of shelf life (fish and fruit)
Semiconductor devices	n-γ flux*	Transistors, diodes, and multifunctional devices
Food sterilization	Co^{60}	Military rations (chicken, beef, pork, and ham)
Enzyme sterilization	Machine	

*Nuclear reactor.

The overall research and development on process radiation includes the following projects:

1. Studies in radiation technology which consist of theoretical and experimental work on basic and applied radiation mechanisms to aid in the development of large-scale radiation chemical processes.

2. Studies on radiation chemical methods for the preparation of fluorinated organic compounds either as an alternative to classical methods of synthesis or of those compounds which are not conveniently prepared by conventional procedures.

3. Induced gelation involving the incorporation of multifunctional monomers into polymers for the purpose of (*a*) reducing the radiation dose required for attaining incipient gelation and high gel contents and (*b*) inducing the radiation

*Such a review has been presented at the conference on "The Application of Large Radiation Sources in Industry," sponsored by the International Atomic Energy Agency in Salzburg, Austria, May 27–31, 1963. Copies of the review paper entitled "Technical Developments in USAEC Process Radiation Development Program," may be obtained upon request to the Division of Isotopes Development, U. S. Atomic Energy Commission, Washington 25, D. C., U. S. A.

crosslinking of polymers that normally are degraded upon irradiation. Crosslinking upgrades the properties of polymers by increasing the molecular weight (viscosity), melting temperature, and tensile strength while decreasing solubility and elongation.

4. Investigation of radiation-induced reactions in swollen polymers, natural and synthetic, to produce novel polymers that apparently cannot be produced by other means.

5. Development of technology leading to the preparation of novel and useful polymers of controlled structure by radiation-induced graft polymerization. The objective is to provide an understanding of the mechanisms involved in radiation-induced graft processes and to create specific polymeric structures of interest to commerce and unique materials for aerospace applications.

6. Radiation-induced gas-phase reactions utilizing Kr^{85}.

7. Development of a chemical dosimetry system wherein rupture of the organometallic bond occurs proportionately to gamma dosage and a solar-cell dosimeter for high-level dosimetry.

Several process radiation systems currently under advanced development within the AEC program are described below in detail.

Wood-Plastic Combinations

Improvement of wood properties has been the objective of many diversified research programs totally apart from nuclear energy development. Among the important areas of study are resistance to decay and insects, dimensional stability, and mechanical properties. Impregnating various woods with monomers and then polymerizing using radiation produces a wood-plastic combination that exhibits superior properties in these areas.

The experimental work on radiation-produced wood-plastic combinations includes:

1. Determination of the properties of various wood-plastic combinations produced by radiation polymerization.

2. Chemical studies to determine *in situ* polymerization rates of various monomer systems, characterization of the polymer, and investigation of methods of accelerating the polymerization to permit reduction of total radiation dose requirements.

3. Development of data necessary to control the quantity of monomer absorbed by various wood species and examination of test specimens to determine uniformity of polymer loading.

Radiation-produced wood-polymer combinations prepared from a variety of woods and monomers exhibit increased compressive strength and hardness. White pine–polymethyl methacrylate, for example, which contains 0.6 part of polymer per part of base wood, has a compressive strength at the proportional limit about 40% greater than that of the unmodified wood. Although higher polymer loadings do not yield further increases in compressive strength, the hardness continues to increase with increasing polymer content. A loading of 1.55 parts of polymer yields a pine product having a hardness (measured by ball penetration) 130% greater in the axial direction, 450% greater in the tangential direction, and 500% greater in the radial direction than unmodified white pine. A specimen containing 0.75 part of polymethyl methacrylate absorbed 40 g of water when immersed for 20 days, whereas a control absorbed 110 g.

Birch and white oak combined with polymethyl methacrylate and polyvinylacetate were as much as 200% harder than unmodified controls, and compressive strength at the proportional limit showed a marked increase. For example, white oak–polymethyl methacrylate containing 0.25 part polymer was 50% stronger than the unmodified control.

In general, wood-plastic combinations exhibit increased hardness and compression strength and absorb water more slowly, thereby reducing changes in dimension due to varying environmental conditions.

Incorporation of a rigid, unyielding material into the wood structure can be expected to increase brittleness. Qualitative observation indicates that this is true in many of the wood-plastic combinations prepared. The testing program is being extended to include measurement of this property.

The rates of radiation polymerization of several monomers impregnated in wood have been measured, and the time required to effect complete polymerization has been determined in some cases. For example, a dose of 6×10^6 rads, applied at the rate of 1.3×10^5 rads/hr, was found to effect complete polymerization of methyl methacrylate in white pine. Various additives are being tested to decrease the total dose requirements. Carbon tetrachloride

allows a considerable decrease in dose when used at concentrations of 10% in styrene and methyl methacrylate. Additives also receiving attention include ethylene glycol dimethacrylate, polyethylene glycol dimethacrylate, tetraisopropyl titanate, and divinyl benzene. Density measurements provide a rapid and accurate method for determining the concentration of polymer formed in the wood. The impregnation conditions necessary to obtain desired loadings of polymer in various wood species are being studied. Reasonably good control can be obtained by properly specifying the vacuum pretreatment and the length of soaking time.

Future work will include:

1. Characterization of the physical properties of additional wood-plastic combinations. The testing program will be expanded to include shear strength, static bending, toughness, and, if possible, decay and insect resistance.
2. Development of additional data on radiation polymerization rates of additives intended to decrease dose requirements.
3. Specification of conditions necessary to effect the desired extent of impregnation of selected species of wood.

Hydrogenation of Coal

Conversion of coal to gasoline and oil products by an economically competitive process would open up a vast new market for the coal industry. Technically such conversion can be accomplished by hydrogenation of coal. One way this is done involves coal extraction to obtain a meltable organic fraction (80 to 85% of coal), followed by hydrogenation over conventional contact catalysts at high hydrogen pressure and elevated temperature, to yield the oil products.

A serious expense in the present technology is replenishment and regeneration of the contact catalysts used for hydrogenation. The catalyst is inactivated by metallic "poisons" in the extract and under some conditions by carbon laydown. Since radiation cannot be diminished by the above factors, radiation catalysis of the hydrogenation step provides sufficient merit to warrant specific investigation. Accordingly, a full research program has been established.

In this research a 90-curie Sr^{90} source is used inside a 10,000-psi 500°C heavy-steel-walled reaction vessel. Pure beta-emitting radioisotopes are preferred over gamma emitters to obtain good absorption efficiency in the reaction vessel. The radiation source was specially designed to withstand the conditions of elevated temperatures and pressures in the reactor, to permit beta irradiation of the reactants, and to avoid radiocontamination of products. This radiation processing unit is operated on batches, with the feed (usually melted-extract–tetralin solutions) pumped into the reaction vessel, held at 5000 psi hydrogen pressure at 380 to 430°C, followed by drainage of the liquid products from the vessel. Appropriate safety precautions such as the contaminant cell, barricades, and remote radiation monitoring are used.

Ten radiation processing test runs have been completed so far, as have comparative nonradiative runs. The residence time for the reactions was 1 hr, with a dose rate of 0.98 Mrad/hr. Results on the coal extract from two runs are given in Table 6.

The first row of data shows the conversion yields and hydrogenation (7.3% hydrogen in feed, raised to 10.37% in products) obtainable with a fresh contact catalyst. The last column shows that hydrogen addition is less in runs with radiation than without any catalyst at all. This may be due in part to inefficient mixing with the radiation source in the vessel. In any case hydrogen addition is not accelerated by radiation in all the runs made. Despite this the yield of desirable oils in the boiling range 100 to 400°C is seen to be somewhat higher for the last radiation run shown than for the noncatalyzed, thermal reaction run. The yield of 250 to 400°C oil seems definitely higher. Tentative explanations are that radiation may be promoting hydrogen transfer from the tetralin solvent or that radiation is promoting selective "cracking" of the extract to the 250 to 400°C oil without much hydrogenation. The third set of data in Table 6 shows that hydrogen gas saturation diminished the yield of 250 to 400°C oil, so that the yield in the last run may also be partially due to less inhibition of the cracking reaction by less hydrogen in solution. Results of radiation runs under the same conditions on the model aromatic compounds biphenyl and methylnaphthalene show no radiation-induced hydrogenation in the liquid phase in excess of that obtained by thermal reaction at 429°C. No tetralin was present in these runs.

Liquid-phase hydrogenation of pure aromatic model compounds is not measurably promoted

Table 6 TYPICAL RESULTS, COAL EXTRACT-TO-OIL CONVERSION

(*T*, 429°C; *P*, 500 psi; Feed, Extract : Tetralin in a 1 : 1 Ratio with 7.3% Hydrogen)

Catalyst	Yield from coal extract, %			H in overall product, %
	100 to 250°C oil	250 to 400°C oil	100 to 400°C oil	
Cobalt, molybdenum, nickel, sulfur	33.47	17.69	51.16	10.37
None (thermal reaction)	5.12	9.65	14.77	8.24
	8.55	9.65	18.20	8.24
Sr^{90} radiation (H_2 saturated)	8.73	6.79	15.52	7.76
Sr^{90} radiation (not H_2 saturated)	4.82	15.80	20.72	8.00
	5.32	16.30	21.62	8.00

in excess of the thermal reaction at elevated temperatures. Likewise, hydrogen addition to coal extract is not promoted by radiation alone. However, some increase in production of desirable oils from coal extract was observed when radiation was used and when the reactants were not saturated with H_2 gas. Since the lack of dissolved H_2 may also have affected this "cracking" oil yield favorably, additional experiments are needed to isolate the effect of radiation alone. Then a more exact conclusion can be made about the specific benefit of radiation.

Radiation runs on coal-extract–tetralin solution without hydrogen gas pressure will be conducted to measure the effect of radiation alone on the hydrogen transfer or cracking production of 250 to 400°C oil from the extract. This can be a very useful selective reaction, which can be extended by increased dose if proved. Test runs with both radiation and disposable homogeneous or contact catalysts will also be made.

Transmutation Doping of Semiconductor Devices

All semiconductor devices, e.g., transistors, diodes, and microelectronic circuits, consist of electronically active impurities (denoted N or P type, depending on whether they contribute Negative electrons or Positive holes) deposited within a semiconductor crystal in patterns of various degrees of spatial complexity. The conventional fabrication processes diffuse these impurities into the crystal from the outside.

In neutron transmutation doping these electronically active impurities are inserted into the semiconductor crystal in a spatial pattern by means of neutron irradiation. A nuclear transmutation reaction occurs in which a semiconductor nucleus (Ge^{70}, for example) absorbs a neutron and is transmuted to a new nucleus (e.g., Ga^{71}, a P type dopant in germanium) which is electronically active in the semiconductor. The spatial pattern of this impurity deposition is controlled by completely enclosing the semiconductor within a "radiation die." This radiation die is a thin boxlike structure composed of thermal-neutron absorbing material, e.g., boron or cadmium, with a pattern of slits to permit neutrons to enter into the semiconductor in a geometry determined by the slit arrangement. The work on this project consists of the detailed development of the technology of the neutron transmutation doping process, an experimental proof of the principle, and the application of this technology in semiconductor device fabrication.

The analyses performed to date indicate that a variety of simple and complex devices, including many with unusual performance characteristics, can be fabricated at reasonable cost by this process. These include diodes, transistors, microelectronic circuits, solar cells, and thermoelectric elements. Doped configurations of unusual and useful shapes can be produced, and wide barrier regions can be made.

The radiation die can be made at low cost and with considerable flexibility when cadmium is used. Silicon carbide devices, which cannot be doped by conventional methods, can be prepared by neutron transmutation doping.

Semiconductor specimens are enclosed in radiation dies with appropriate slit patterns. These assemblies are then "canned" and irradiated. The resistivities and other appropriate electrical characteristics are measured before

irradiation, and suitable surface treatment and passivation are performed. After irradiation the specimens decay (for about two months) until their activation is negligible. They are then annealed to remove gamma-radiation dosage effects. Dislocation density is measured by microscopic examination of surface-etching effects, both before and after annealing. The specimens are then tested for resistivity and for their device performance characteristics to determine if the predicted device performance can be obtained. Devices to be fabricated include diodes, transistors, solar-cell elements, and microelectronic circuits of germanium and silicon.

Analyses and calculations have been performed to determine the effects of radiation-die parameters, slit geometry, neutron flux levels, and reactor location parameters on the characteristics of the device produced. Neutron irradiation will be carried out in the Oak Ridge Research Reactor. Work is now in progress on the fabrication of the radiation dies, the irradiation assembly, and specimen preparation.

The principal results to date are:

1. Design data have been developed for predicting the device characteristics from the process parameters, e.g., radiation die and slit characteristics and neutron flux levels.

2. Data on fabrication methods for the radiation die have been obtained. Both boron and cadmium can be suitably fabricated into radiation dies with the required characteristics. Cadmium is particularly convenient since it is easy to fabricate, and its poorer epithermal shielding characteristics can be circumvented by appropriate design.

3. Heat-transfer experiments have shown that the semiconductor temperature can be held below 250°C during the irradiation process if the die holder and die are fabricated to close tolerances. Impurity diffusion is thereby avoided.

The first specimens should be ready for irradiation soon. A number of experiments are planned for proof-of-principle and to fabricate and test a number of progressively more complex devices (including test of their performance characteristics) in order to demonstrate the utility of this process.

Polymerization of Ethylene

During the past decade several studies have been made on the Co^{60} gamma-radiation-induced polymerization of ethylene. Because of the potential importance of a Co^{60}-based process, additional work has been initiated to increase the knowledge of the behavior of this system in a radiation field.

The present research program is concerned with measurement of the reaction yield and rate of ethylene polymerization in a Co^{60} gamma field. These data were obtained by utilizing a continuous pressure-recording technique. The conditions under which the rate was measured varied in pressure from less than 68 to 680 atm and in temperature from −78 to 40°C. Polymerization takes place in the liquid phase at −78°C at a very low rate. In the gas phase, at 20 to 40°C, a strong inhibition and retardation effect due to oxygen impurity was noted. The induction period as a function of oxygen concentration was determined. The experiments indicate that the effect of the presence of polymer on the polymerization rate is smaller than the effect of oxygen and ethylene pressure. The differential G values, based on monomers alone, were determined, and attempts were made at expressing these values as a function of ethylene pressure and radiation intensity. In the range of 80 to 350 atm and 21,000 to 132,000 rads/hr, the rate is proportional to the 0.93 to 1.13 power of the pressure and to the 0.33 to 0.46 power of the intensity in the falling pressure period. At 133,000 rads/hr, a minimum differential G value of 15,000 was obtained. At 21,000 rads/hr, a maximum value of 47,000 was reached. Additions of solid substrates of alumina and molecular sieves increase the G value by a factor of 2.2 to 2.5 and the rate by a factor of 1.6 to 1.9, indicating a surface effect.

All the polymer produced in this work by gas-phase radiolysis was a fine white powder. In the homogeneous experiments the entire vessel was filled with polymer. The polymer could be removed from the vessel in a coherent mass and had various bulk densities depending on the percentage conversion. Under a microscope the polymer powder showed agglomerates of submicron particles.

Because of the varying pressures during the formation of the mass, it is not possible to obtain a quantitative evaluation of the polymer as a function of pressure. However, qualitative effects can be obtained.

1. The density measurements, falling in the range 0.93 to 0.94 g/cm^3, indicate a medium-

density polymer. Annealed measurements show values of about 0.95 g/cm^3. Amorphous polymer has a density of 0.86 and fully crystalline material, 0.99 g/cm^3.

2. The reduced viscosity measurements indicate that the molecular weight increases with decreasing total dose. The average molecular weight for polymers formed up to 250 atm falls in the range 80,000 to 300,000. For the liquid-phase polymer, the intrinsic viscosity measurements indicate that the material is of low molecular weight.

3. The addition of argon decreases the molecular weight.

4. The degree of branching, indicated by the number of methyl groups per thousand carbon atoms, decreases with increasing pressure, decreasing dose rate, and decreasing total dose. The range of values for initial pressure of 650 atm is indicative of medium-density high-pressure conventional material.

5. The polymers that were fused before removal from the vessel were crosslinked with the exception of those produced at low total doses, which tend to be noncrosslinked. Most of the polymers removed from the vessel without melting showed a high degree of solubility in decalin and indicate less than one crosslink per molecule. It appears that the solid polymer contains residual radical species, which result in crosslinking on heating. Exposure of the unreacted radicals to the atmosphere, on the other hand, can cause termination of radicals due to oxidation.

6. The melt flow and melt index indicate the processibility of the polymer. The radiation-produced polymer has a lower melt-flow index and a higher melt index than the low-density polymer. The melt index for conventional high-pressure polyethylene is of the order of 1.7, and the melt flow for high molding is about 50 to 60.

7. The material has a high crystalline melting point.

8. The decalin-soluble noncrosslinked polymer could be made into a pliable film by hot pressing.

9. Low melt-flow index, low branching, and noncrosslinked materials are indicative of a higher molecular weight linear polymer. The type of polymer produced is dependent on combined conditions of temperature, pressure, intensity, and total dose.

Further characterization and evaluation of the polymer await experiments at constant pressure, both in static and in flowing systems. Work is continuing to determine the quantitative effect of the various parameters on the rate of polymerization.

Textile Modification

Under this program, textile processes in two general areas have been investigated. These include the use of various nuclear radiations to modify fiber properties and the use of graft copolymerization to modify the chemical structure of fibers. The results of three and one-half years of research have shown that the most promising field for commercial application is in the area of fiber modification by radiation-induced graft copolymerization. Means of overcoming fiber deficiencies and thus producing materials of more functional or aesthetic value by this procedure are being investigated.

New uses for old fibers will be found, and fibers will be upgraded in their performance characteristics. In the work to date, emphasis has been placed on procedures for getting satisfactory grafting at levels of radiation where fiber deterioration due to exposure to the radiation itself is minimized. The approach was to expose a number of different textile fibers to vinyl monomers in the vapor phase, using a third component in the system to accelerate the grafting process. The vapor-phase technique gave grafted products of good uniformity, and for this reason work was largely confined to this system.

The fibrous materials that are the least stable to radiation are the ones that were grafted most readily. Thus the best results were obtained with cellulosics, cellulose esters, polyamides, and polypropylene. Polyesters and acrylics showed little tendency to graft. The best vinyl monomers investigated were acrylonitrile, methyl acrylate, ethyl acrylate, and butadiene. The presence of water in the system was necessary for the grafting of all these monomers to the cellulosics and the polyamides except the cellulose esters and polypropylene. Other factors that affected the rate of grafting were the concentration of the monomers in the vapor phase, the presence of oxygen, and the temperature of the system. Generally, the amount of grafting increased with increased monomer content; oxygen had an inhibiting

effect, presumably by acting as a scavenger for free radicals; and the effect of temperature was to decrease the amount of graft at any given radiation dose.

Fibers of the different chemical types were partially evaluated after grafting with four different levels of the four vinyl monomers mentioned. Stress-strain properties, moisture regain, solubility, softening point, and weather resistance of a few selected materials were determined. The cellulose ester grafted with acrylonitrile behaved somewhat differently than the other fiber types. Its breaking strength was unchanged as a result of grafting, but the elongation of the fiber decreased. This resulted in an increase in modulus. Moreover, the yield point was increased as the amount of graft copolymerization increased. The overall data indicated that cellulose esters were cross-linked as well as grafted by acrylonitrile. Generally speaking, the other fiber types showed a slight deterioration in physical properties when grafted with the different monomers, although in no case was the deterioration severe. The moisture regain of all fiber types with all monomers was decreased. The solubility of the grafted materials depended on the monomer used. The addition of acrylonitrile tended to insolubilize all fiber types in their common solvents. Greater solubilities were generally obtained when ethyl acrylate was used as the monomer. One level of grafting for each monomer on each fiber type resulted in insolubility. The addition of the monomers to the thermoplastic fibers failed to increase their softening or melting point except in the case of acrylonitrile. The addition of this monomer to a cellulose ester and to polypropylene gave a material that charred without melting. Cotton grafted with as little as 3.25% acrylonitrile was highly resistant to cellulose-degrading microorganisms. The addition of styrene to nylon appeared to improve weather resistance.

Concurrently with the above work, experiments were initiated to correlate both the kinetics and diffusion for the vapor-phase technique on grafting acrylonitrile to cellulose acetate, nylon, polypropylene, and polyester fibers. The kinetics of polymerization and the rate of diffusion of monomer both play a very significant part in this process, and accurate data are necessary for the optimum design of graft-polymerization processes. Reaction velocity constants were determined for propagation, termination, and reaction with oxygen. The diffusion of acrylonitrile monomer did not obey a simple mechanism.

Future work will involve continued studies of the kinetics and of the role of diffusion of monomers into the fiber substrates using computer techniques. The evaluation of important textile-fiber properties will be continued. Some preliminary work has indicated the possibility that cotton cellulose might be made more dimensionally stable by the use of a radiation process.

Radiation Pasteurization of Foods

Research on radiation preservation of food is being carried out in many countries, including the United States and Japan as well as a number of countries throughout Europe. Major developments have been achieved during the past year which contribute to the steady advance on radiation preservation of foods technology. Potatoes irradiated to 10,000 rads for control of sprouting have been approved for human consumption in Canada. Last fall more than one million pounds of potatoes were irradiated in Canada as part of a field "test" program to establish better potato response to radiation treatment, process and storage conditions, and economic and consumer acceptance. The Russian Ministry of Public Health has reportedly authorized the use of irradiated potatoes by consumers, and an experimental production facility to irradiate up to 25 tons of potatoes per year is planned.

In the United States radiation-sterilized bacon was approved for human consumption by the Food and Drug Administration in February 1963. A formal petition for clearance of wheat and wheat products treated with radiation for disinfestation purposes has been submitted to the Food and Drug Administration.

The U. S. Army Radiation Laboratory, Natick, Mass., was completed in 1962 and used for studies on radiation sterilization of food products important to the military establishment. The facility was designed and constructed by the AEC for the Department of Army. The Laboratory contains the world's largest Co^{60} radiation source (1.3 million curies), a high-energy 24-Mev 18-kw linear accelerator, and a complete food preparation area.

The AEC research program on radiation pasteurization of foods aims at developing the technology for providing fresh-like fish and fruits far from harvest areas, thereby opening up new markets for these products. Relatively low radiation doses (less than 500 kilorads) are being used to inhibit bacterial growth for periods ranging from days up to several weeks under normal refrigerated distribution and storage conditions for perishable foods. The low-dose radiation processing of foods program of the AEC is complementary to the radiation sterilization of foods program of the Department of Army where high doses of radiation are used to completely destroy bacteria and enzymes and permit food storage without refrigeration for months.

The AEC research and development program is limited to selected fruit and fishery products and emphasizes factors such as storage and shelf-life extension, treatment variables (including radiation dosage), quality (including organoleptic effects), wholesomeness, and packaging materials studies. In addition, continuing consideration is being given to marketing and consumer-acceptance studies. The selected food items under study are:

Fruit: strawberries, citrus, peaches, grapes, pears, cherries, and tomatoes. (Preliminary experiments have been carried out on figs, pineapples, papayas, asparagus, and nectarines.)

Fish: haddock fillets, soft-shelled clam meats, Pacific crab meat, petrale sole fillets, and shrimp. Recent work has been initiated on cod, pollack, and ocean perch; however, results are not available for reporting at this time.

Radiation for control of sprout inhibition, disinfestation of grain, and alleviation of quarantine problems is also being considered.

Research results achieved to date with both fruits and fishery products show continuing promise for successful development of radiation pasteurization technology and for ultimate commercialization of the process.

Progress on specific fruit products:

Strawberries: The amount of decayed fruit after a simulated transit and marketing period is considerably less in fruits irradiated at doses of 200 kilorads than in the control lots. The texture of the irradiated fruit is altered less than that of other fruits. This derives in part from the fact that strawberries, both irradiated and control, become progressively firmer in cold storage. Taste panel studies reveal that irradiated strawberries are not significantly different in flavor compared to nonirradiated fruits. Color, aroma, and texture are significantly different in irradiated fruits but are not objectionable to the taste. Ascorbic acid levels in irradiated strawberries are not altered by a nutritionally significant amount.

Lemons: Severe injury symptoms were found in irradiated lemons when the fruit was stored for periods of one month or more. Air pockets develop in the fruits adjacent to the lobe membranes. Ascorbic acid content declines by 50% in fruits subjected to 200 kilorads, and the "buttons" are destroyed, making the fruit very susceptible to *Alternaria citri*.

Oranges: This species does not show the adverse effects observed with lemons. Air pockets do not develop, the ascorbic acid level in irradiated fruit is not significantly different from that of the controls, and the "buttons" are not adversely affected. There is a change in flavor at 200 kilorads with increasing storage time, but, with two months' storage, this changed flavor is acceptable.

Peaches and Nectarines: The capacity of these fruits to ripen is not altered by irradiation. They are subject to dramatic effects by radiation on the texture of the flesh. Some varieties of these species have shown a drop in flesh firmness of 70 to 85% at 200 kilorads.

Grapes: White varieties of grapes are more susceptible to adverse radiation effects than are red varieties. Maturity of fruit has a profound effect on its response to radiation. As the fruit becomes riper, the adverse effects of radiation are less. The Thompson seedless variety shows moderate to severe textural changes at doses that may prove effective against *Botrytis* and *Rhizopus*.

Bartlett Pears: Ripening of this fruit is clearly inhibited at doses of 250 kilorads and higher. There is an immediate softening of the flesh at doses as low as 100 kilorads, and at 300 kilorads the firmness of the flesh is only 50% of that of the unirradiated fruit. Subsequent to the initial decrease in firmness, fruits irradiated at 250 kilorads and higher soften more slowly than the control lots. Fruits subjected to ripening-inhibiting levels of radiation in air are "mealy" when ripe and do not develop full yellow color. Irradiated fruits respond more slowly to ethylene treatment than do control fruits.

When pears were irradiated in a nitrogen atmosphere, their color development, texture, and flavor were more nearly normal, indicating that the adverse effects noted above may be an indirect effect of high ozone concentration in the fruits rather than an effect of radiation per se.

There is a spurt of ethylene production by irradiated pear fruits immediately after treatment. Thereafter ethylene production by fruits subjected to 300 and 400 kilorads is much less than that of unirradiated fruits. Fruits subjected to 100 and 200 kilorads produce considerably more ethylene than do the control fruits, suggesting that with this species there may be a very narrow range of doses between adverse and nonharmful effects on ripening by irradiation.

Sweet Cherries: Cherries have been studied for the past three years. During this time casual observation and tasting of the fruit have given promise that these fruits can well tolerate radiation doses up to 250 kilorads. It is also reasonable to assume that such a dose would result in an important extension of the postharvest life.

Tomatoes: No retardation of ripening has been found at doses below 200 kilorads. Doses between 200 and 400 kilorads retarded ripening and decay development, but this range of dosage gave an undesirable effect on flavor.

Figs: Mission figs seem to respond favorably to radiation. Although no inoculations were made, irradiated fruit showed much less decay than did control lots. There were no apparent ill effects of doses as high as 300 kilorads.

Pineapples: These fruits respond very favorably to radiation. At 200 and 300 kilorads, there is a slight bronzing of the exterior color of the fruits. Flavor of the irradiated fruits seems the same or slightly improved over that of control fruits.

Papayas: Fruits were shipped by air from Hawaii and subjected to doses of 200 and 300 kilorads. The fruits seemed to ripen more slowly than unirradiated fruits. There was no evidence of adverse effect on flavor.

Asparagus: The postharvest elongation of asparagus spears was reduced by 66% and tip curvature by 80% at doses as low as 5 kilorads. There were no adverse effects on quality of the product at doses as high as 150 kilorads.

Progress on specific fisheries products:

Haddock Fillets and Soft-Shelled Clam Meat: Research results show conclusively that, at the optimum radiation dose levels of 250,000 and 450,000 rads, respectively, haddock fillets and soft-shelled clam meats can be kept satisfactorily for 30 days at 33°F. At these optimum dosage levels, no noticeable "irradiation" odor is detectable in the products. An increase in storage temperatures from 33 to 42°F decreased shelf life by about 50%. Results have not yet demonstrated a difference in product quality due to use of vacuum or air packaging.

Pacific Crab Meat: Radiation pasteurization of Pacific crab meat packed in cans increases the storage life at ice temperatures by five times, or from 7 to 35 days. Radiation dose levels of 200 to 350 kilorads appear to be the optimum. Quality of the product remains excellent.

Petrale Sole Fillets: 400 kilorads appears to be the optimum radiation dose. A sevenfold extension of shelf life, 5 to 35 days, was attained at 33°F in the vacuum-packed samples. The storage life of this product was half as great at 42°F as compared to 33°F. In air-pack samples, processed under the same conditions, an off-flavor, described as rancid, developed.

Shrimp: The shelf life of radiation-processed shrimp, held at 33°F and using a radiation dose of 100 kilorads, was doubled from 20 to 40 days. At this dose level, organoleptic and chemical results were consistently better than those of the unirradiated samples.

Cooked irradiated shrimp appear to have excellent keeping qualities as shown by bacteriological, chemical, and organoleptic tests.

A valuable finding is that "black spot" (a melanosis resulting from the action of phenol oxidase in shrimp) is virtually eliminated in radiation-processed products.

Design and Fabrication of Irradiation Facilities

Three research irradiators have been completed. Each utilizes approximately 30,000 curies of Co^{60} with a specific activity ranging from 7 to 18 curies/g. These irradiators are designed with a built-in flexibility to permit the performance of irradiations under various types of atmospheres, as well as under a temperature variation from 10 to 150°F. The gamma flux for each has been measured to be about 2.5×10^5 rads/hr, and the irradiation chamber is capable of handling a package measuring $14 \times 18 \times 6$ in. Transportable irra-

diators are designed for use in semiproduction-scale acceptability, storage, and shipping tests. The transportable irradiator provides a radiation processing capability close to the harvest areas as well as data on process parameters and economics.

Design concepts for two transportable irradiators have been completed. One will use a 100-kilocurie Co^{60} radiation source, and the other, a 2-Mev 3-kw electron machine radiation source. These irradiators have a designed minimum production rate of 500 lb/hr at a radiation dose of 250 kilorads.

The Marine Products Development Irradiator is designed to demonstrate the technical and economic feasibility of radiation pasteurization of fishery products on a near-commercial scale. It will use a 235,000-kilocurie Co^{60} source and will have a production rate of 1000 lb/hr at 500 kilorads. The facility will be located at Gloucester, Mass., which is the center of the east-coast fish-packing industry in the United States. The facility will be operated for the AEC by the Department of the Interior, Bureau of Commercial Fisheries. Construction on the Marine Products Development Irradiator was scheduled to begin in the summer of 1963, and it is expected to be completed in July 1964.

Radioisotope Technology Developments

This phase of the Isotopes Development Program is concerned with creating new and improved technology leading to beneficial applications in areas such as water exploration and management, atmospheric pollution, meteorology, oceanography, and crime detection. Research and development are being carried out on problems involving neutron activation analysis, radiometric analysis, process control with low-level tracers, radiation absorptiometry, ionization technology, nondestructive testing by radiography, and isotope measurement systems. Emphasis in the research and development work is directed primarily to filling critical gaps in technology which are acting as major deterrents to advancement of the use of radioisotopes.

Although important contributions have been made in each of the above-mentioned areas, discussion will be limited to work of most timely interest and significance.

Neutron Activation Analysis

Neutron activation has moved out of the research laboratory into broad application throughout science and industry in the United States. Some 25 private organizations now offer neutron activation as a commercial service. The method is beginning to find wide use in the chemical and petroleum industries.

The development of new instrumentation, particularly in neutron generators, multichannel analyzers, and detectors, continues to accelerate. The rapid developments in instrumentation have opened up new areas for the investigator, especially in the utilization of fast-neutron reactions. This has revealed gaps in the basic knowledge which must be filled.

The AEC is continuing its strong interest in activation analysis by supporting a broad range of research and development projects. Fundamental studies are being carried out on gamma-ray spectra of neutron-deficient nuclides as well as on solid-state detector systems which will provide superior beta and low-energy photon resolution as compared to crystal detectors. Development of instrumentation systems to provide rapid, automatic analyses of very large numbers of samples is an important area of work, as is development of improved neutron sources. Much effort is being placed on applying neutron activation analysis to problems of major national importance, including analysis of environments in space and in oceanography.

Analysis of the Lunar Surface. The AEC and the National Aeronautics and Space Administration are jointly sponsoring a program to develop a workable, remote, activation analysis technique for elemental analysis of the surface of the moon. It is planned that a neutron source and detector will be included as part of the instrument package of a scientific lunar landing mission. A small area of the surface of the moon will be neutron irradiated, the induced gamma radiations will be detected, and the data will be telemetered to earth. The data will then be analyzed by an automatic computer-coupled system that will identify the major chemical elements in the moon surface sample.

A working system has been operated successfully under simulated remote conditions. The activator is a neutron tube that produces a flux of 5×10^8 neutrons/(cm^2)(sec) at the target located at a distance of about 0.1 in.

from the end of the accelerator unit. This flux is barely adequate, and a more powerful source is being considered. The detector unit is a 3- × 3-in. NaI(Tl) scintillation crystal, a photomultiplier tube, a preamp, and a lead collimator. The control unit powers the accelerator, controls pressure automatically, and programs the analysis sequence. This involves activation by the neutron source for 5 min, rotation of the assembly to place the detector over the activated sample for 10 min, and transmission of the data signals to the earth-stationed multichannel analyzer.

Tests have been conducted with simulated moon matrices using a mixture of aluminum oxide (Al_2O_3), ferric oxide (Fe_2O_3), magnesium oxide (MgO), and silicon dioxide (SiO_2). The gamma radiation was analyzed with a 400-channel transistorized analyzer, and photopeak counts for the various components were taken from a plot of channel number vs. activity. Spectrum-stripping techniques were used to resolve peaks containing activity from more than one radioactive source.

Data from an actual moon shot are to be processed with the Mark II automatic gamma analyzer, on command from the moon probe. This analyzer is a modification of the Mark I, previously reported at this conference in 1961. Advances in Mark II include faster operation by direct coupling of computer tape, elimination of some interfering elements by a "dynamic subtract" unit, and improved resolution of gamma peaks.

Shipboard Neutron Activation Analyzer. The shipboard analyzer performs rapid elemental analysis on core samples of the ocean floor on the vessel as they are collected. This aids effective exploration by making it possible for the ship to modify its survey pattern in accordance with the trends indicated by the data.

The unit was developed in conjunction with Lamont Geological Laboratory of Columbia University. The special neutron source, a small, rugged (d,T) neutron generator which emits 10^8 neutrons/sec, is contained in a lead and paraffin shield designed to permit insertion of an entire ocean-bottom core sample so that intervals along the core length may be activated without cutting the core. A shielded 3- × 3-in. NaI(Tl) scintillation crystal is used to observe the activity induced in the core, and the gamma spectrum is analyzed in a 400-channel analyzer. The analysis of cores for their content of silicon, aluminum, magnesium, sodium, and iron is made by neutron activation, and for their content of potassium, uranium, and thorium, from the spectra of their natural radioactivities. The activation conditions are: 10 min for core irradiation, 1 min for transfer of the core to the detector, and 10 min for measurement of the induced activity. Analyses of standard samples have indicated the following sensitivities, in mg/cm^3: aluminum and silicon, 0.3; magnesium, 1.6; sodium, 2.2; and iron, 3.8.

Coal Analysis and Grading. In cooperation with research groups of the coal industry, the AEC is sponsoring investigations into the feasibility of using activation analysis to continuously measure the British thermal units and ash content of coal. By the use of activation analysis using a neutron generator emitting 14.5-Mev neutrons, combined with the related techniques of inelastic scatter and capture gamma-ray analysis, it appears to be possible to analyze for the elements that determine the British thermal unit value of coal, i.e., carbon, nitrogen, oxygen, and hydrogen; the water content; the sulfur content; and ash. The data for the several quantities are fed into a computer for automatic calculation of the British thermal units. It is anticipated that such units can be installed at conveyor belt lines carrying the finished coal. These can be coupled to automatic sorting equipment.

Carbon content is determined to an estimated accuracy of ± 1% using inelastic scattering of fast neutrons. Oxygen content is determined to about ± 2.5% using neutron activation analysis. The ash constituents, silicon, aluminum, and iron, are determined by activation to within about ± 5%. Methods for sulfur, nitrogen, and hydrogen are under development.

Automated Activation Analysis in Metabolic and Environmental Studies. Determination of the role of trace elements in metabolism and its correlation with the incidence of disease and existent chronic deficiency conditions are of major interest to medical research in the United States. The analytical problem in this research is one of tremendous scope inasmuch as a serious study should include analysis for several elements in enough samples to investigate statistically the major environments across the nation and to establish the trace-element content of all the life-sustaining por-

tions of these individual environments. This includes the atmosphere, the drinking water, and the food supply. Ideally, seasonal changes should also be investigated. Further, any specific effect in a particular area, such as coal dust in a coal-mining region, for example, must be covered intensively. Clearly, such a program poses a monumental analytical problem, involving hundreds of thousands of samples.

Previous studies of trace-element effect on life processes have been limited to specific elements and usually to specific geographic areas where the effect of a particular deficiency or excess was conspicuously apparent. Studies of fluoride effect on teeth and toxic selenium effects are examples.

Neutron activation has provided the first general analytical approach that appears to be economically capable of coping with the enormous task. Automated computer-coupled analysis in environmental health studies is now under way under joint sponsorship of the National Institutes of Health and the AEC. Elements selected for initial studies are silica, selenium, and tellurium. The neutron activation technique is particularly advantageous for determination of selenium. This is a difficult analysis by classical methods, involving considerable risk of losing selenium in the ashing process. The selenium activation technique is nondestructive, rapid, and sufficiently sensitive.

Activation Analysis in Law Enforcement. Activation analysis is particularly suited to the chemical analysis of material evidence taken in law-enforcement investigations because it is nondestructive and the sensitivity is adequate. The AEC has been conducting extensive studies in this area, both at its Oak Ridge National Laboratory and at private laboratories. Much of the work at ORNL has been conducted in close consultation with the U. S. Department of the Treasury. It has included such diverse problems as identification of automobile paint, soot, drugs, soil samples, hair, and fingernails.

Compendium of Activation Analysis. Under the AEC program, continuing attention has been given to providing a standard reference compilation of information on neutron activation analysis. The publication of standard reference atlases of gamma spectra has been completed. A compendium on neutron activation analysis, which is a companion to the standard gamma atlases, is nearing completion at ORNL. The compendium provides the analytical chemist with needed information for determining the pertinence of activation to his particular problem (with the standard spectra atlases) and most of the information needed to begin work in activation analysis.

Although the procedures detailed in such a reference are quickly outdated, the modification or amplification of the basic material is much slower. Supplements will report significant developments as they achieve the status appropriate for inclusion in a reference work.

High-Altitude Atmospheric Density Sonde

A direct-measuring balloon sonde to telemeter density information during flight has been developed in cooperation with the U. S. Weather Bureau and the U. S. Air Force. This sonde is intended to replace the indirect and frequently inaccurate calculation of density based on thermistor data.

The device is a forward-scatter gas-density gage using a 10-mc gaseous Kr^{85} source. It has been flight-tested under Air Force supervision. Use of the forward-scatter geometry, which gives a better signal-to-noise ratio than backscatter geometry, and a 14-in. gap between source and detector, provides sensitivity for measurement in the extremely rarefied atmosphere up to 140,000 ft. The average density error in the interval from 45,000 to 140,000 ft appears to be $\pm 0.05\%$. The instrument contains two scintillation detectors (one to measure and subtract background), a battery high-voltage supply, two amplifiers, a difference circuit, and a radio transmitter.

Deep-Water Isotopic Current Analyzer (DWICA)

One of the many problems confronting both the hydrologist and the oceanographer has been the lack of a suitable method to measure precisely and to record accurately direction and flow of slow-moving currents in the range 0.01 to 1 knot. Now, for the first time, such a method has been developed, based on an isotopic principle.

The principle of the isotopic technique depends on injection of a radioisotope into a moving water environment and measuring drift as the radioactivity passes a detector. In this system a series of 16 detectors is arranged

in a wagon-wheel configuration with a diameter of 2 ft. The detectors completely encompass a radioisotope injection mechanism located in the center of the configuration so that any release of activity will be seen by the detector system. After the radioisotope is injected, the direction and rate are determined by time of arrival of the radioactivity at a particular detector in the configuration.

The isotope method is far superior to previously available techniques for determining current direction and flow rates.

The greatest advantage of this new system is that there are no moving parts to impose friction drag on the system as occurs in the case of a rotor mounted on a sapphire bearing. No matter how well a rotative cup is designed, there will always be a certain amount of friction, whereas with the isotope method the actual drift of the current is the mechanism by which the radioactive material is transported and recorded.

The first field tests of this instrument were made on Dec. 14 and 15, 1961, in Round Bay, Md., in 19 ft of water. The instrument was placed 13 in. off the bottom, and measurements were made with I^{131}. The tests began at 5:00 p.m. on Dec. 14 and ran until 3:00 p.m. on the 15th. During these 22 hr, measurements were made at intervals of from 6 to 20 min.

A second series of field tests was performed in the same location on Dec. 19 and 20. Measurements were made from 5:30 p.m. on Dec. 19 until 3:00 p.m. on the 20th, at intervals of 1 to 20 min.

The results of the first series of measurements in Round Bay are shown in Fig. 1. In this figure direction is given by the upper curve; note that more than one full turn of the compass is plotted. Current speed is indicated by the lower curve. Current directions are indicated on the left-hand axis, and speeds, on the right-hand axis. Between 5:00 and 10:00 p.m. on Dec. 14, the current direction rotated in a clockwise fashion through 360°. For the next 10 hr, the current direction remained upstream, about 340°. However, starting about 8:00 a.m. on Dec. 15, the current direction again made a complete clockwise turn about the compass in approximately 3 hr. Current direction was downstream for the final 3 hr of the measurements.

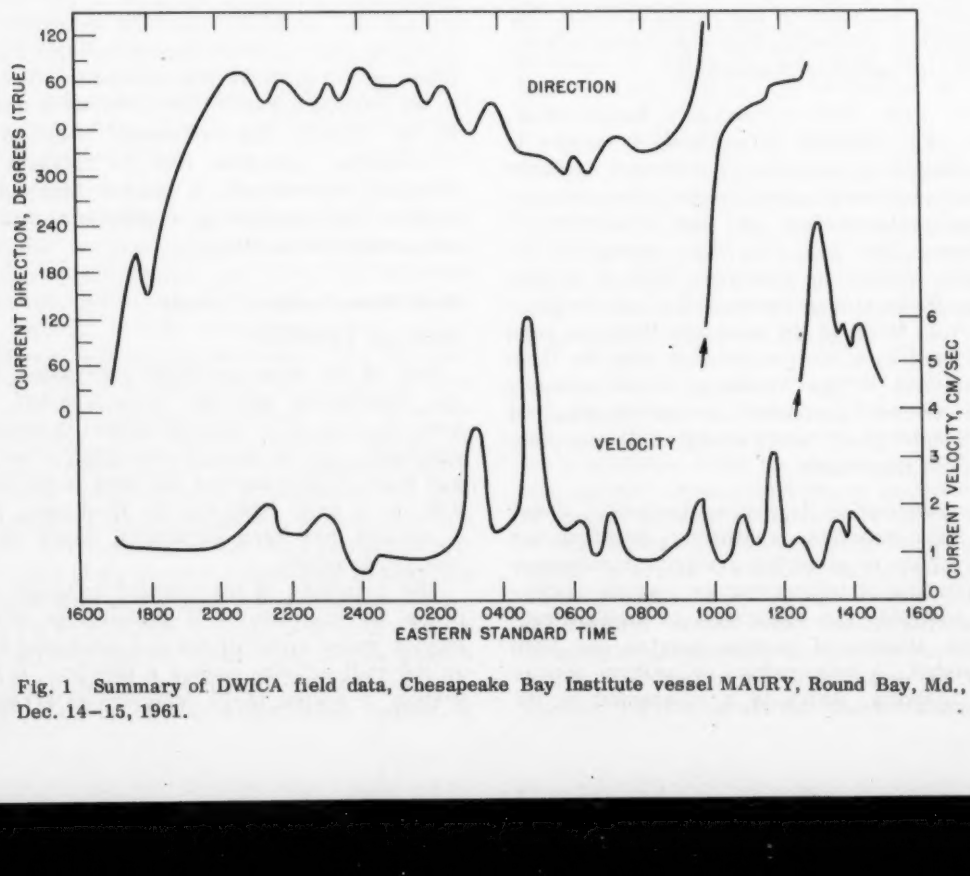

Fig. 1 Summary of DWICA field data, Chesapeake Bay Institute vessel MAURY, Round Bay, Md., Dec. 14–15, 1961.

The results show that current movement upstream has a longer duration than current movement downstream. Further, the data indicate a correlation of the movement with flood and ebb tide, which was predicted by experts of the Chesapeake Bay Institute. However, until these tests were made, there was no method of verification.

This test represented the first successful attempt to log currents with velocities as low as 0.01 knot.

The instrument has been modernized with computer-logic circuits to make it completely automatic. The instrument consists of the following units: (1) an electronic servo-injector control system which automatically adjusts the amount of radioactivity injected at the start of current velocity measurements and makes the necessary corrections *in situ* when the velocity increases or decreases; (2) an automatic digital converter system which prints out the following information: date and time of measurement, drift time of injected activity, and the number of the detectors that observed the activity (i.e., direction); and (3) an automatic program control system which determines the intervals of measurement. This, of course, depends on the velocity of the current being measured. The present design of the DWICA can measure velocities in the range 0.001 to 10 knots and has been used by the Chesapeake Bay Institute to collect data on summer and winter cycles in Chesapeake Bay. The lower limit of the instrument is determined by molecular diffusion of the radioactive material in still water.

The instrument is being redesigned and pressurized to measure ocean currents at 6000-ft depths. The new instrument will include many of the above-mentioned features, as well as several additional ones, i.e., a compass for proper orientation at great depths and a unit that can generate its own radioisotopes *in situ*. The generator will produce In^{116} by activation of an indium solution with ten 1-curie Am^{241}-beryllium neutron sources with a total flux greater than 10^5 neutrons/(cm^2)(sec).

Table 7 COMPARISON OF TRITIUM LABELING METHODS*

Tagged compound	Wilzbach method: Treatment at 25°C, hr	Wilzbach method: Specific activity of purified tracer, mc/g	New method, $TH_2PO_4 \cdot BF_3$: Treatment Hr	New method, $TH_2PO_4 \cdot BF_3$: Treatment °C	New method, $TH_2PO_4 \cdot BF_3$: Specific activity of purified tracer, mc/g
Benzene	200	60	2	65	514
Toluene	200	90	4	25	393
Tetralin	200	148	6	25	240
Naphthalene	200	242	2	65	440
Decalin	200	156	8	25	12

*Starting material consists of 10 curies of tritium in either case, as gas or reagent. All samples to be labeled weighed 5 g.

The first field test of the redesigned instrument is scheduled for August 1963 and will be conducted in cooperation with the U. S. Navy Oceanographic Office.

New Improved Tritiation Method for High-Molecular-Weight Organic Compounds

High-molecular-weight hydrocarbons are now successfully labeled by a new tritium reagent. This new technique produces labeled compounds of a higher specific activity than produced by the radiation-induced self-labeling method of Wilzbach. It is faster and cleaner and requires less initial tritium to produce a product of higher specific activity. The principle of this new technique is based on the hydrogen-tritium exchange brought about in the presence of a tritiated acid catalyst.

The tritiating reagent is a complex of tritiated phosphoric acid and boron trifluoride. It normally has the formula $TH_2PO_4 \cdot BF_3$, but the tritium-to-hydrogen ratio is arbitrary since the complex is easily prepared by stoichiometric admixture of tritiated water and phosphorus pentoxide followed by saturation with boron trifluoride.

Its applicability is best illustrated in Table 7, which compares this new technique and the standard radiation-induced self-labeling one.

Table 8 DEGREE OF LABELING IN SIX HOURS COMPARED TO ULTIMATE THEORETICAL LIMIT (T = 23°C)

Compound	Theoretical limit of tagging, μc/g	Tagging in 6 hr, μc/g	Ultimate tagging achieved in 6 hr, %	Experimentally observed limit of tagging, μc/g
Benzene	630	108.2	17.2	~630
Decalin	435	31.4	7.2	
Tetralin	545	542	99.4	~550
Toluene	603	554	92.0	~560
Methylcyclohexane	381	9.36	4.1	
Naphthalene	2830	1290	45.6	
Cyclohexane	No exchange tagging			

Many advantages are seen from the comparison. Experimental studies of radiochemical purity and tritium activity in the tracers show that the $TH_2PO_4 \cdot BF_3$ reagent produces no highly tagged side products, thus obviating extensive purification as required in the radiation-induced method. The method is rapid, and pure, usable tracers are often prepared in a few hours as compared to days by the Wilzbach method. High-specific-activity compounds, up to curies per gram, can be produced when desired by starting with a large tritium content in the reagent. Table 8 shows the theoretical limit reached in a 6-hr test at 23°C.

As seen in Table 8, labeling takes place on contact under ambient conditions and causes tritium to exchange readily with hydrogen atoms on all aromatic positions and on tertiary carbon atoms; there is no labeling on nonbranched aliphatic carbon atoms.

Kryptonates as Universal Radioactive Indicators

Development of a general, or "universal," radioactive indicator technique, called "kryptonate," which was first reported at this conference in 1961, has been completely successful. Krypton gas containing 5% of the radioactive isotope is incorporated into the substance by either bombardment with ionized krypton or diffusion under high temperature and pressure. The ionization procedure, which was originally favored, is now used only for selected substances that cannot withstand the pressures or temperatures required of the more efficient diffusion process.

Typical conditions for the diffusion saturation of a substance with krypton are temperatures around 650°C and pressures to 5000 psi. Several hours to three days are required. The saturation activity that can be attained is a function of the structure of the compound. Substances with numerous interstices of the proper dimensions to contain the krypton atom assume the highest saturation activity. Pyrolytic graphite has been saturated to an activity of 1 curie/g, whereas with less favorable structures the activity is frequently limited to millicuries or microcuries per gram. The diffusion method has successfully induced some measurable, though often small, permanent krypton activity in all substances tested so far. Therefore the title universal indicator is still justified.

Uniform incorporation of krypton into a solid is possible if the substance is kryptonated in the form of a fine powder that can be subsequently compressed into a solid. The temperature stability of the kryptonates varies. Some lose most of their activity at atmospheric pressure when the temperature is elevated only 75 or 100 degrees, whereas kryptonated copper metal retains 75% of its activity to 900°C. Generally, the stability at room temperature is excellent.

The possible applications of the kryptonates in science and technology range from pure chemistry to applied mechanical engineering. One of the most important applications is the accurate measurement of atmospheric moisture, especially at high altitudes. Kryptonates appear useful here, as Kr^{85} incorporated into desiccant material is released when these substances absorb water vapor. A research study based on the formation of hydrates to release Kr^{85} and another based on chemical reaction to form the hydroxide, thus releasing Kr^{85}, are in progress.

Section

I

Isotopes and Radiation Technology

Introduction to Isotope Technology

In this first issue a brief general review of the principles of isotope production and utilization will be followed by more detailed consideration of various applications in a number of areas. In subsequent issues advances in technology and new applications will be covered, but, in general, routine applications will be ignored. Since there are only certain utilization principles, we hope that novel disclosures in one area may suggest analogous techniques to users in other areas.

A chronological outline of the development of the isotopes program is included to provide background information. Many publications, including annotated bibliographies, may be used by those who want more detailed information on early work and developments to supplement the reviews in the first issues of this quarterly. Among these are the three-, five-, and eight-year summaries of isotope distribution and utilization;[1a–1c] the subsequent bibliographies published by the U. S. Atomic Energy Commission (AEC);[2a–2g] the list of special sources of isotope information;[3] the reports of the AEC to Congress;[4] and various AEC and nongovernment publications.[5a–5f]

Only about 100 radioisotopes are now routinely available,[6] but more than 1000 have been isolated and more than 1500 identified. Certainly many of these have properties suitable for a wide variety of applications. We plan to keep readers abreast of progress in this direction, particularly as enriched targets and higher fluxes are used, and to suggest profitable areas of utilization. The subject "Stable Isotopes" will be covered in a separate section.

That the program has grown rapidly is illustrated by examination of information on shipments, authorizations, licenses, and publications (Table I-1).

Table I-1 GROWTH OF ISOTOPES PROGRAM

To (date)	Total shipments	Total authorizations and licenses	Cumulative total publications
August 1949	11,400	5,000*	1,850
June 1951	18,900	13,100*	3,000
December 1954	64,200	37,155*	7,000‡
December 1958	118,600	4,347	15,000‡
December 1962	170,000	5,881†	40,000‡

*Until 1956 individual authorizations were processed for each isotope. After that, users were able to obtain licenses that could authorize any number of isotopes.

†The 1962 figure does not include licenses transferred to the jurisdiction of California (580), Kentucky (68), Mississippi (37), and New York (637).

‡Estimated on basis of sampling from *Nuclear Science Abstracts*, *Chemical Abstracts*, and *Journal of Nuclear Medicine*.

Table I-2 NUMBER OF LABELED COMPOUNDS AVAILABLE IN 1949–1962

Isotope	1949*	1962†
S^{35}	5	131
I^{131}	5	56
P^{32}	13	81
C^{14}	~100	~1300

*Page 68 of Ref. 1a.

†Pages 13–132 of Ref. 7.

ping, licensing, and publications since 1946 (Table I-1) and by comparison of the number of labeled compounds available in 1949 with the number available in 1962 (Table I-2).

Isotope Production

Any applications involving radioisotopes presume availability of suitable nuclides. Most radioisotopes are prepared by neutron irradiation in reactors, by proton or deuteron reactions in cyclotrons, or by separation of fission products that remain after the processing of spent fuels (Tables I-3 to I-5).

Table I-3 NEUTRON REACTIONS

Reaction	Example
(n,γ)	$Co^{59}(n,\gamma)Co^{60}$
	$Fe^{58}(n,\gamma)Fe^{59}$
(n,p)	$N^{14}(n,p)C^{14}$
	$S^{32}(n,p)P^{32}$
(n,α)	$Ca^{40}(n,\alpha)Ar^{37}$
	$Li^{6}(n,\alpha)H^{3}$
$(n,\gamma;EC)$	$Xe^{124}(n,\gamma)Xe^{125} \xrightarrow{EC} I^{125}$
$(n,\gamma;\beta^-)$	$Pd^{110}(n,\gamma)Pd^{111} \xrightarrow{\beta^-} Ag^{111}$

Table I-4 CYCLOTRON REACTIONS

Reaction	Example
(p,n)	$Li^{7}(p,n)Be^{7}$
(d,n)	$Ge^{72}(d,n)As^{73}$
$(d,2n)$	$Cr^{52}(d,2n)Mn^{52}$
(d,α)	$Mg^{24}(d,\alpha)Na^{22}$
(p,pn)	$Ca^{48}(p,pn)Ca^{47}$
(p,α)	$Mg^{25}(p,\alpha)Na^{22}$

Table I-5 FISSION PRODUCTS

Isotope	Fission yield, %	Half-life, years
Kr^{85}	0.3	10.3
Sr^{90}	5.9	28
Tc^{99}	6.1	2×10^5
Ce^{144}	6.1	0.8
Pm^{147}	2.6	2.6
Cs^{137}	5.9	30
I^{129}	1.0	1.7×10^7

Isotope Utilization

The principles of isotope utilization have been reduced to four major types, or methods of use, as given below.

Effects of Radiation on Materials

The radioisotope is used as a fixed source of radiation, comparable to ordinary radium or X-ray machines, and the radiation is allowed to impinge upon the material that is to be affected by the radiation. There are many important examples of this application. The target material may be a plastic, exposed in order to alter its properties (Fig. I-1); a patient with a malignancy, receiving therapy; food being irradiated to retard decay (Fig. I-2); or a phosphor being excited to luminescence by an admixed isotope.

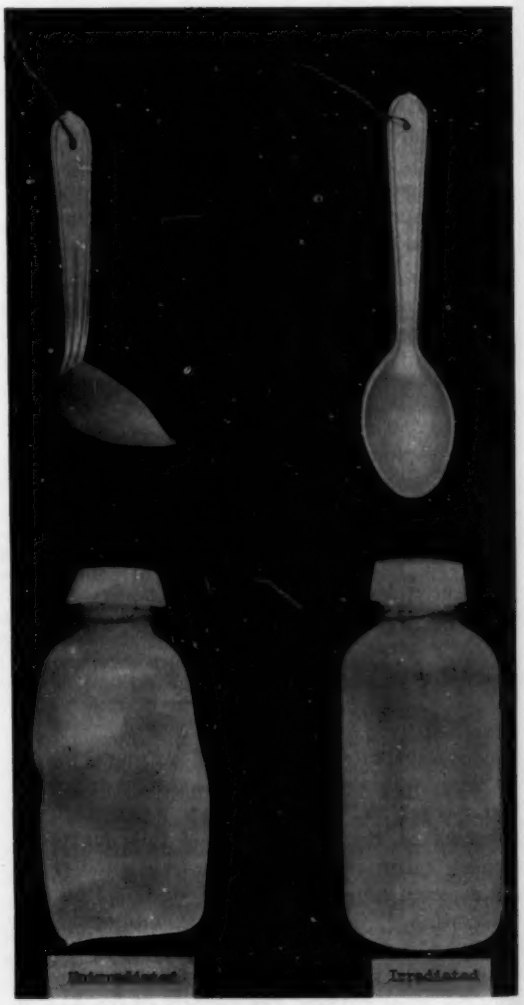

Fig. I-1 Effect of radiation on thermal stability of polyethylene. (Courtesy of Battelle Memorial Institute.)

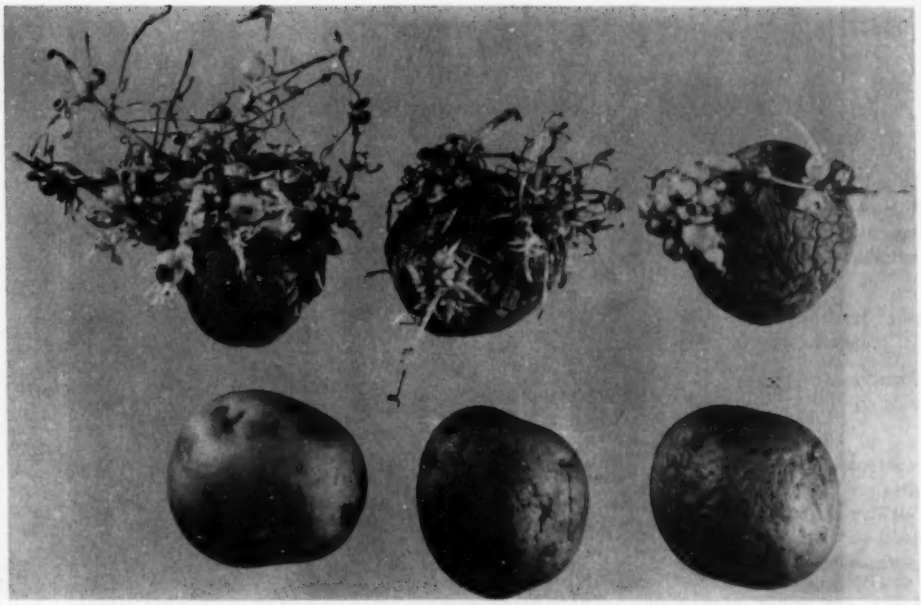

Fig. I-2 Effect of gamma radiation on sprouting and softening of potatoes; dosages varied from 0 (top left) to 106,000 r (bottom left). (Courtesy of Brookhaven National Laboratory.)

Effects of Materials on Radiation

Here the target material affects the radiation in such a way as to provide information about the material. The radiation, either reflected from or penetrating the material, is measured by a counter or photographic emulsion, depending on the information required. For example, in industrial or medical radiography, a radioactive source can replace X-ray machines to photograph bones, teeth, or castings. Industrial utilization includes many important applications to process and product control: measuring blind (inaccessible) liquid levels (Fig. I-3); gaging thickness of moving sheets of paper, plastic, and metal (Fig. I-4); controlling cigarette densities; radiographing castings and internal structures of equipment; and, in some of the more recent applications, using the isotopic heat resulting from the complete absorption of the radiant energy by a heavy metal such as lead or uranium (Fig. I-5).

Tracing of Radioactive Atoms

Perhaps the most interesting and significant use of radioisotopes takes advantage of the continuous "broadcast signal" generated by the decaying radioisotopes. Suitable detectors can be used to follow even the most complicated courses of these atoms as they take part in chemical reactions or biological processes or as they are involved in physical transfer. The radioisotope tracer may be used to study surface phenomena such as induced passivity; it may be incorporated in a compound added to water running through a leaking pipe; it may be used to trace oil flow in pipelines; it may be an active atom in sugar (e.g., C^{14}) for studying body metabolism; or it may be a label in a fertilizer compound used to study the mechanism of fertilizer uptake.

Isotopic Heat

The heat generated by isotopes during radioactive decay has recently been used to generate electricity. Although current methods for the conversion of heat to electricity are not highly efficient, devices of this kind are being used as power sources in space and in remote regions of the earth (Fig. I-6). The first use of an isotopic heat source in space was on Transit 4A, a navigational aid placed in orbit June 29, 1961. The Pa^{238}-powered generator in this satellite operated two of the four radiotransmitters.

Fig. I-3 Determination of hidden liquid level in cans. (Courtesy of Industrial Nucleonics.)

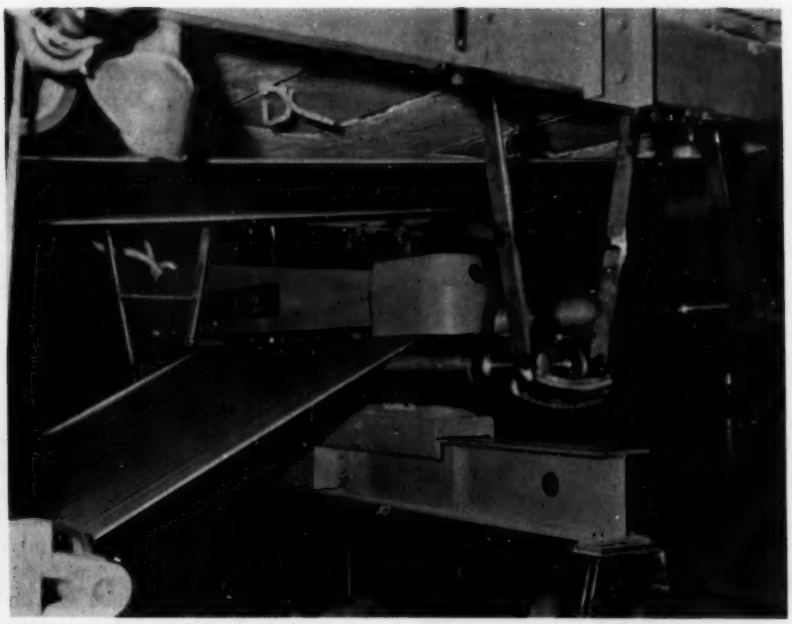

Fig. I-4 Gaging the thickness of a moving sheet of gummed paper. (Courtesy of Industrial Nucleonics.)

Fig. I-5 Cutaway view of 250-kilocurie $SrTiO_3$ thermoelectric generator for operating a shore light at Curtis Bay, Md. (Courtesy of Martin-Marietta Corp.)

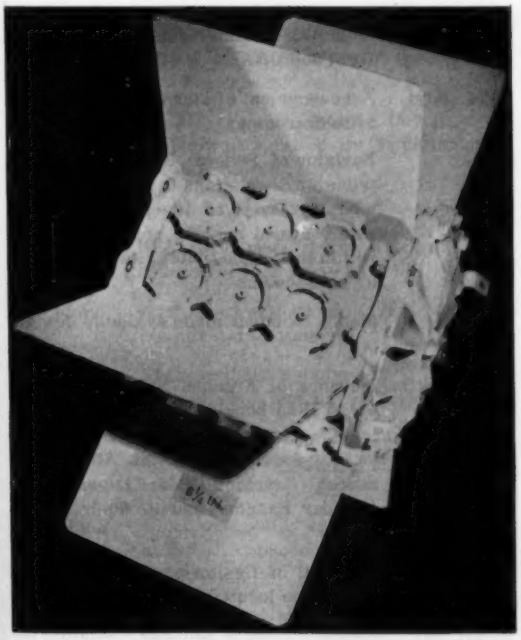

Fig. I-6 SNAP-9 device for use in forthcoming space shots.

Chronological Outline of Development of Isotopes Program

1946 (June)	First public announcement of reactor-produced radioisotopes available under the distribution program appeared in *Science* (served as 1st radioisotope catalog)
1946 (Aug.)	First reactor-produced radioisotope shipment from Oak Ridge
1947 (Jan.)	Jurisdiction of program turned over to civilian Atomic Energy Commission (AEC)
1947 (Mar.)	Initiation of reactor service irradiations; i.e., a service whereby the radioisotope user may submit his own sample for irradiation or neutron bombardment in a nuclear reactor
1947 (July)	First compound labeled with radiocarbon (methyl alcohol) available from the Commission
1947 (Sept.)	Routine availability of chemically processed radioisotopes with standard specifications
	Initiation of a program for international distribution of radioisotopes
1948 (Jan.)	Formation of an Advisory Committee on Isotope Distribution (to replace interim policy committee set up under the Manhattan Project)
	Initiation of a program for distribution on a loan basis of more than 100 varieties of electromagnetically concentrated isotopes of about 30 elements
	Publication of regulations by the Interstate Commerce Commission for shipment of radioisotopes by common carrier (rail and truck)
1948 (Apr.)	Initiation of a program making available free of production costs three isotopes — radiosodium, radiophosphorus, and radioiodine — for research, diagnosis, and therapy of cancer and allied diseases
1948 (June)	Opening of first Commission-sponsored training course in radioisotope techniques by Oak Ridge Institute of Nuclear Studies
1948 (July)	Beginning of production and distribution of radioisotope-labeled compounds by commercial firms

1948 (Sept.) Distribution of tritium (radioactive hydrogen) and stable He^3

1948 (Sept.) Completion of a nationwide intercomparison of I^{131} radiation analyses conducted by the National Bureau of Standards (NBS) and AEC

1949 (Feb.) Extension of the Commission's program for support of cancer research to include the availability of all normally distributed radioisotopes free of production costs for use in cancer research

1949 (June) Initiation of a program for cyclotron production and distribution of certain long-lived radioisotopes not producible in the nuclear reactor

1949 (July) Enactment of regulations by the Civil Aeronautics Board for shipment of radioisotopes by commercial aircraft

1950 (Mar.) Completion of new large-scale radioisotope processing facilities at Oak Ridge National Laboratory (ORNL)

1950 (Oct.) Announcement of program for returning radioactive wastes to Commission facilities for disposal

1950 (Dec.) Initiation of cooperative visitation with state health departments to radioisotope users

1951 (Jan.) Announcement that Brookhaven National Laboratory (BNL) will provide reactor irradiation services and radioisotopes

1951 (Mar.) Elimination of requirement for publication of results obtained with AEC-produced radioisotopes

1951 (Apr.) Publication of first issue of the quarterly bulletin, "Isotopics — Announcements of the Isotopes Division"

Publication in *Federal Register* of regulations governing radioisotope distribution, including designation of "generally licensed" quantities (requiring no license application)

1951 (July) Expansion of international distribution of radioisotopes to include all reactor-produced radioisotopes (except tritium and Po^{210}) and to authorize the export of these materials for use in scientific research, medical research, industrial research, medical therapy, and industrial applications

1951 (Nov.) First radioisotope (I^{131}) accepted by Federal Food and Drug Administration as effective new drug; P^{32} accepted June 1952; and colloidal Au^{198} accepted October 1954

1952 (Jan.) First film of thirteen-part radioisotope training film series, "The Radioisotope," made available for distribution

1952 (Apr.) Announcement of availability of neutron activation analysis service at ORNL

1952 (May) Publication of AEC-financed supplement to the *Journal of the American Chemical Society* on labeled-compound synthesis

1952 (June) Revision of AEC cancer program to make most radioisotopes available at 20% of catalog prices

1952 (July) First authorization of industrial waste-disposal service

1953 (Dec.) Publication in the *Federal Register* of criteria governing the authorization of AEC-supplied radioisotopes for research and development, for human use, and for use under a general authorization; use of general-license quantities of radioisotopes in humans expressly prohibited

1954 (Jan.) Announcement of availability of facilities for irradiation of large items at BNL

Publication by USAEC of list of 492 isotope-labeled compounds available from 13 commercial suppliers, the NBS, and ORNL

1954 (July) Announcement of standard reactor units of radioisotopes

Revision of Federal Radioisotope Distribution Regulations to cover radioisotopes produced in any publicly or privately owned nuclear reactors located within the United States, its territories, or possessions

Stable isotopes made available for export

1954 (Sept.) Issuance of Argonne National Laboratory (ANL) booklet, "Irradiation Services," containing detailed information on services available at ANL's Research Reactor CP-5, Experimental Breeder Reactor, and its 60-in. cyclotron

1954 (Oct.) Approval of fission-product separation plant to be located at ORNL

Announcement of gamma irradiation facilities at National Reactor Testing Station to provide gamma-ray fields of the order of 10 million roentgens per hour

1955 (July) Large-scale production of high-specific-activity Co^{60} begun at AEC's Savannah River Plant

1955 (Nov.) AEC withdraws from processing and distribution of cyclotron-produced isotopes

1956 (Feb.) Generally licensed quantities of radioisotopes increased, permitting wider use and interchange of small samples without an application; simplification of international distribution of radioisotopes permitting U. S. licensees to export radioisotopes of elements with atomic numbers 3 to 83

Sale of enriched stable isotopes approved by AEC

1956 (Aug.) Records relating to radioisotope licenses made publicly available in AEC's Public Document Room

1958 (Mar.) Cobalt-60 encapsulation services turned over by AEC to private enterprise

1958 (Apr.) Fission-product Ce^{144} reduced by AEC to $1.00/curie for orders of 100 kilocuries or more

1958 (Aug.) Fission-product separation plant started operation at ORNL

1958 (Oct.) Isotope Development Program initiated for "encouraging the growth and development of radioisotope and radiation uses in industry"

1959 (Jan.) AEC Division of Isotopes Development Advisory Committee on Radioisotopes and Radiation formed

1959 (Aug.) Routine availability of Ca^{47} announced

1960 (May) AEC withdraws from sale of Po^{210} sources of less than 20 curies

1960 (July) AEC discontinues providing standard reactor-irradiated units

AEC reaffirms policy of withdrawal from neutron irradiation service

Abbott Laboratories announces availability for first time of I^{131} from private manufacturer

1961 (Jan.) Electromagnetic separations facilities at ORNL expanded from 4 to 28 calutrons

1961 (May) Cobalt-60 metal sources become available from private industry; AEC limits supply from Commission facilities to orders of 100 kilocuries or more; large-order price reduced to $1.00/curie for specific activities less than 30 curies/g

1961 (June) Completion by ORNL of first large fission-product source for use in thermoelectric generator—17 kilocuries of $SrTiO_3$, 110 thermal watts; later placed in generator developed by Martin Company to power an unmanned arctic weather station for the U. S. Weather Bureau

1961 (July) AEC terminates its radioisotope discount program in medicine, agriculture, and biology research

1961 (Aug.) Routine availability of I^{125} announced

1961 (Nov.) AEC withdraws Mound Laboratory as a supplier of plutonium-beryllium neutron sources

1961 (early) First private production and marketing of Co^{60} (General Electric and Westinghouse)

1962 (early) First private production and marketing of short-lived isotope (Isoserve, Inc.)

1962 (Mar.) AEC withdraws from neutron activation analysis service in favor of industry

1962 High Intensity Radiation Development Laboratory to generate the engineering design and operational knowledge essential to expansion of the potentials of process radiation completed at BNL

1962 (Mar.) Fission-product Cs^{137} reduced by AEC to $0.50/curie for orders of 100 kilocuries or more

AEC makes up to 10 g of Am^{241} and Np^{237} available to industrial licensees

Fission-product Sr^{90} reduced by AEC to $0.75/curie for orders greater than 30,000 curies

1962 (May) Completion by ORNL of first large gamma-ray (Cs^{137}) source for thermoelectric generator—27 kilocuries of cesium borosilicate, 125 thermal watts; later inserted in generator by Royal Research Corp. for operation of a deep-sea seismograph station for Lamont Geological Observatory

1961–1962 Fabrication of a series of SNAP sources for Navy navigational beacon, Coast Guard buoy, Navy antarctic weather station, and unmanned Navy floating weather station; sources by ORNL, generator assembly by Martin Company; 31 to 225 kilocuries of $SrTiO_3$, 195 to 1400 thermal watts

1962 (Sept.)	First AEC license to industry for land disposal of by-product wastes
1962 (Oct.)	AEC withdraws from fabrication and sale of Po^{210} sources of less than 1 kilocurie for private use
1962 (Dec.)	Development of Cs^{137}–lithium silicate glass at ORNL for BNL large-scale research gamma irradiator
1963 (Jan.)	First Sr^{90} fuel source processed by private industry
	Price reduction for Co^{60} in the high-curie-supply area; aimed at encouraging greater commercial reprocessing and distribution
	AEC withdraws from providing Ir^{192} (solutions and sealed sources) due to industry capability
1959–1962	AEC Division of Isotopes Development arranged and participated in 29 isotope symposia throughout the country to encourage industrial use of isotopes
1962–1963	AEC relinquishes certain of its regulatory authority to states of Kentucky, California, Mississippi, New York, Texas, and Arkansas
1963 (Aug.)	AEC withdraws from by-product waste disposal for licensees

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Section

II

Isotopes and Radiation Technology

Isotope Production and Development

Radioisotopes become available in a number of ways. Megacuries of activity result from the fissioning of reactor fuels; kilocuries of such isotopes as Co^{60} and Ir^{192} and curies or millicuries of many others can be made by activating targets in reactors; and small amounts of a wide variety of nuclides result from the bombardment of suitable targets in accelerators such as cyclotrons.

Stable isotopes are separated by several methods, the most important of which are electromagnetic separation, thermal diffusion, and gaseous diffusion.

In this first issue of *Isotopes and Radiation Technology*, fission-product separation and stable-isotope research and development are discussed briefly. Cyclotron-produced isotopes will be discussed in future issues, and additional details of these processes and material on reactor-produced isotopes will be covered.

Fission Products

When U^{235} undergoes fission, the fission fragments and their decay products comprise a rather sizable percentage of the known isotopic species. Table II-1 lists some of the important radioactive products. Inherent in the operation of all nuclear reactors are the problems of fuel recovery and fission-product waste handling. At present liquid wastes are stored in underground tanks, and solid wastes are buried. However, the economics of waste management are clearly tied in with fission-product separation and recovery.[1] An important experiment involves the Fission Products Development Laboratory (FPDL) at Oak Ridge National Laboratory (ORNL), which was designed specifically to study the feasibility of recovery of kilocurie amounts of potentially useful fission products and fabricate them into various chemical and physical forms for radioisotope applications development work. At first, wastes from both ORNL and the Idaho Chemical Processing Plant were used. More recently, Hanford Atomic Products Operation (HAPO) has been supplying the bulk of the fission products as separated species or groups (e.g., rare earths) for further processing in the FPDL. At present the operations[2] involve primarily separation and purification of Cs^{137}, Ce^{144}, and Pm^{147}. Strontium-90 is received as a purified feed for conversion to appropriate source compounds.

Fission-Product Processing at Hanford

Strontium is recovered from the acid waste raffinate from the Purex plant by precipitation, followed by solvent extraction. Strontium and rare earths are first removed from the solution by a lead-carrier sulfate precipitation at *p*H 1.0, the strontium being carried with the lead sulfate as strontium sulfate. Tartaric acid is added to complex the iron. After centrifugation, precipitated lead, strontium, and rare-earth sulfates are metathesized to carbonates with sodium carbonate, and the carbonates are dissolved in dilute nitric acid. Figure II-1 indicates the basic steps of this process.

The nitric acid solution is now treated with oxalic acid to precipitate lead and rare earths, leaving strontium in solution. Strontium is again precipitated as carbonate to decrease the volume of feed to the solvent-extraction process.

Strontium is purified by solvent extraction with di-2-ethylhexyl phosphoric acid (D2EHPA) as the organic extractant. The final product[3] is reported to be "virtually free of all radiochemical impurities," and cation chemical impurities less than 5% are typical.

The present Hanford Cs^{137} recovery process is outlined in Fig. II-2. The alkaline supernatant liquid is withdrawn from the aged Purex

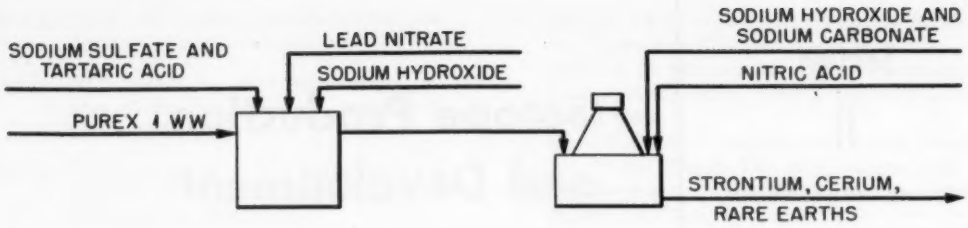


Fig. II-1 Lead-carrier sulfate precipitation.

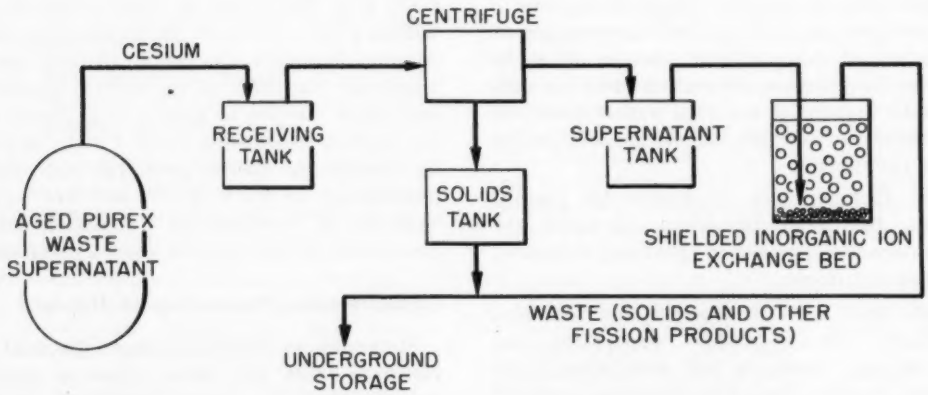


Fig. II-2 Cesium-137 recovery.

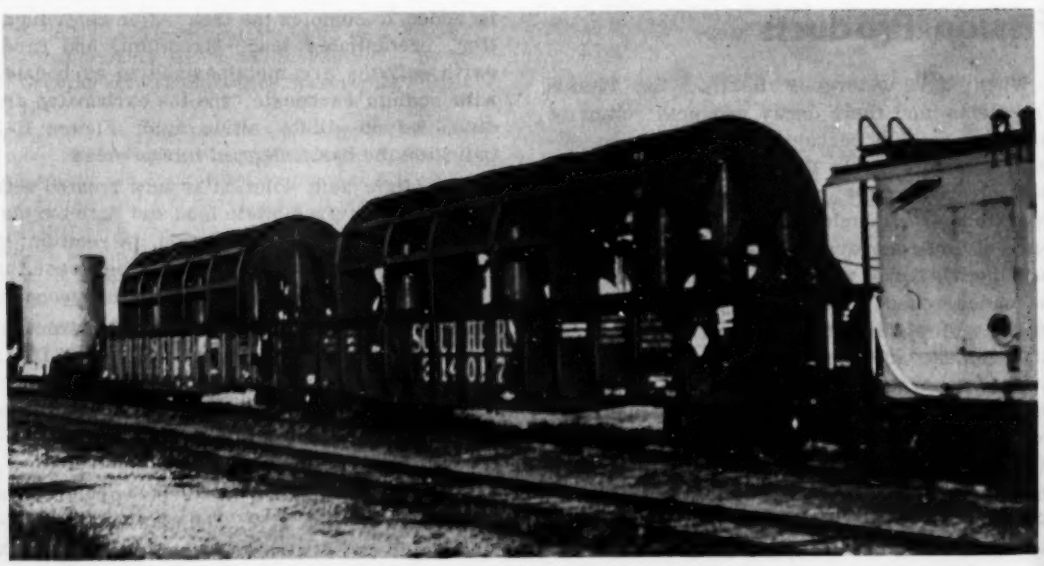


Fig. II-3 "Portable pipeline" for transferring feed.

Table II-1 RADIOACTIVE FISSION-PRODUCT ISOTOPES OF HALF-LIFE GREATER THAN 57 DAYS

Isotope Z	Isotope A	Fission yield, %	Half-life	Thermal-neutron cross section, barns
34	Se^{79}	0.04	6.5×10^4 years	
36	Kr^{85}	0.3	10.27 years	<15
37	Rb^{87}	2.7	6.2×10^{10} years	0.12
38	Sr^{90}	5.9	28 years	1.0
39	Y^{91}	5.9	58 days	1.07
40	Zr^{95}	6.4	63 days	
	Zr^{93}	6.5	1.1×10^6 years	1.4
41	Nb^{93m}	2.1	4.2 years	1.0
43	Tc^{99}	6.1	2.1×10^5 years	19
44	Ru^{106}	0.38	1.0 years	
46	Pd^{107}	0.2	7.5×10^6 years	
47	Ag^{110m}	2×10^{-7}	270 days	
48	Cd^{113m}	?	5.1 years	
49	In^{115}	0.0099	6×10^{14} years	145 + 52
50	Sn^{110m}	0.01	275 days	
	Sn^{123}	0.0012	131 days	
51	Sb^{125}	0.023	2.7 years	
52	Te^{130}	2.0	$>2.0 \times 10^{15}$ years	0.22 + 0.008
	Te^{127m}	0.035	105 days	
53	I^{129}	0.8	1.7×10^7 years	31
55	Cs^{135}	6.2	3.0×10^6 years	10.3
	Cs^{137}	5.9	30 years	< 2
58	Ce^{144}	6.1	290 days	
60	Nd^{144}	6.1	1.5×10^{15} years	5
	Nd^{150}	0.74	2.0×10^{15} years	3.0
61	Pm^{147}	2.6	2.6 years	~60
62	Sm^{147}	2.6	1.4×10^{11} years	
	Sm^{151}	0.5	73 years	12,400
63	Eu^{155}	0.031	1.7 years	14,000

waste that has been stored in tanks, and, after further centrifugation, it is passed through a shielded tank filled with an aluminosilicate ion-exchange material (Decalso). The cesium is sorbed on the inorganic ion exchanger in a form suitable for shipment.

Transportation

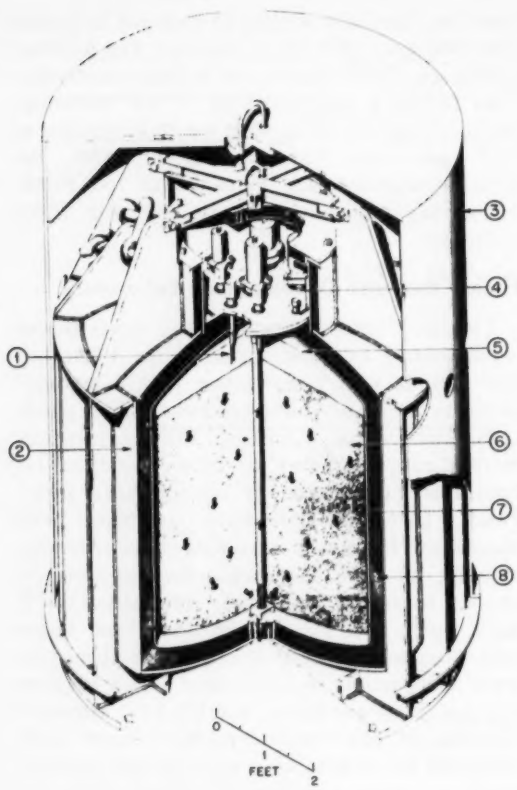
Figure II-3 shows the "portable pipeline" used for transferring feed from HAPO to ORNL. Each gondola carries two 500-gal shielded transfer tanks (STT's) containing 50 kilocuries of Cs^{137} per cask sorbed on a Decalso ion-exchange bed. The flat bed on the extreme left contains a HAPO filter slurry cask with 140 kilocuries of $Sr^{90}CO_3$ powder. A HAPO carrier can safely contain up to 250 kilocuries of $Sr^{90}CO_3$ for shipment as a dried powder on a metal filter.

Figure II-4 is a cutaway drawing of an STT carrier showing its strengthened containment features. The cask weighs 18 tons and is loaded with 400 gal (2200 lb) of Decalso. The limiting factor on Cs^{137} shipments in this particular cask is that it has only 4 in. of lead shielding, which restricts the loading to 50 kilocuries of Cs^{137} per cask. Upon arrival at ORNL, the fission products are unloaded in the FPDL by eluting the ion-exchange bed with HNO_3 solution.

Fission Products Development Laboratory

Fission-product research and development work leading to construction of the FPDL began in 1948 with investigation of a number of methods for removing long-lived fission products from waste solutions. Fission products were first separated in curie quantities in laboratory glassware by ion-exchange processes. Later hundred-curie quantities were separated in small stainless-steel vessels. Finally, in 1958 the FPDL facility went on stream to separate kilocurie amounts of Cs^{137}, Sr^{90}, Ce^{144}, and Pm^{147} from Redox and Purex type wastes. The basic precipitation processes were successfully demonstrated during the first two years of operations, and the first kilocurie amounts of pure fission products were made available for radioisotope applications development. In order to meet the requirements for preparation of large-scale heat sources during the past two years, concentrated, purified, separated fission-product fractions have been shipped from HAPO to the FPDL for further processing on a megacurie-per-year basis.

Figure II-5 is a cutaway drawing of the FPDL with its manipulator cells, wet chemical process cells, analytical cell, tank farm, and associated equipment. The plant is designed for complete processing—from a liquid waste stream to an encapsulated solid product that can be shipped directly to the user. Seven high-level manipulator cells used for product purification and encapsulation contain equipment for precipitation, filtration, calcination, blending, weighing, pelletization, sintering, and welding. Two low-activity-level manipulator cells are available for sampling fission-product solutions and process-control techniques. Eight wet chemical process cells are equipped with stainless-steel piping and vessels to permit a variety of unit operations such as precipitation, centrifugation, filtration, crystallization, decantation, evaporation, and solvent extraction.



1 Thermostatic Level Probes
2 $^3/_4$-In.-Thick Mild Steel Outer Wall
3 Fire Shield
4 16-Gage Stainless-Steel Liner
5 Water Level
6 Decalso Ion Exchange Medium (Sodium Aluminosilicate)
7 $^3/_8$-In.-Thick Stainless-Steel Inner Liner
8 $3^1/_2$-In.-Thick Lead Shielding

Fig. II-4 Cutaway drawing of shielded transfer tank model II (loaded weight, 38,800 lb).

A megacurie-level storage system for Cs^{137} and Sr^{90} was recently installed in the FPDL.

Production. During the past two years, FPDL operations have been concentrated on a two-product stream of Cs^{137} and Sr^{90}. The feed material was obtained from HAPO and then purified to product form. Cesium-137 chloride production exceeded 250 kilocuries, and a 27-kilocurie cesium glass source was produced from this material. The U. S. Weather Bureau 17-kilocurie $SrTiO_3$ source was produced as a prototype in the fiscal year 1961, and the program proceeded rapidly to the production of the 224-kilocurie SNAP-7D source for the Navy.[4] Scale-up from 17 to 224 kilocuries required additional man-days of labor but no changes in the operating procedure. Step-up to a 1-megacurie source can be accomplished at the FPDL with no changes to the facility or to its handling procedures. Flow sheets for Sr^{90}, Cs^{137}, and Ce^{144} are presented in Figs. II-6 to II-8, respectively.

Strontium-90. The product of the Sr^{90} process is $SrTiO_3$, which contains 30 to 35 curies of Sr^{90} per gram of $SrTiO_3$. It is stored in 10-kilocurie lots in stainless-steel containers partially immersed in water for cooling in a storage well. When needed, the dry $Sr^{90}TiO_3$ is formed into pellets and sintered at 1400°C to yield pellets 0.25 to 3.5 in. in diameter. The final pellets, with densities of 3.5 to 4.0 g/cm^3 and activity concentrations well in excess of 100 curies/cm^3, are encapsulated in welded stainless-steel or other metal containers.

The current feed material for the process (highly purified $Sr^{90}CO_3$) is obtained from HAPO. The transported $Sr^{90}CO_3$ is removed from the HAPO shipping casks by water slurrying, followed by dilute nitric acid rinsing. This material is more than 99% radiochemically pure and contains a few percent calcium and barium. The slurry is treated with HNO_3 to yield a $Sr^{90}(NO_3)_2$ feed solution. The FPDL has a storage capacity for 3 megacuries of such solution.

Strontium carbonate is precipitated, in 40-kilocurie batches, with ammonium carbonate in the presence of 5 to 10% excess solid TiO_2. The precipitate is filtered off, and the $SrCO_3$–TiO_2 mixture is calcined at 1100°C to yield $SrTiO_3$. The Sr^{90} content of the product depends on the amount of excess TiO_2 and the concentration of other alkaline earths in the feed.

Strontium-90 can be converted to $Sr^{90}TiO_3$ at a rate of 100 kilocuries per week, the annual production being dependent on the availability of feed material. The preparation rate for pellets varies with the pellet size, encapsulation method, and number of pellets of a given type.

Cesium-137. Assay of the usual $Cs^{137}Cl$ product from the FPDL shows approximately 26 curies of Cs^{137} per gram of CsCl, compared to a calculated activity content of pure ma-

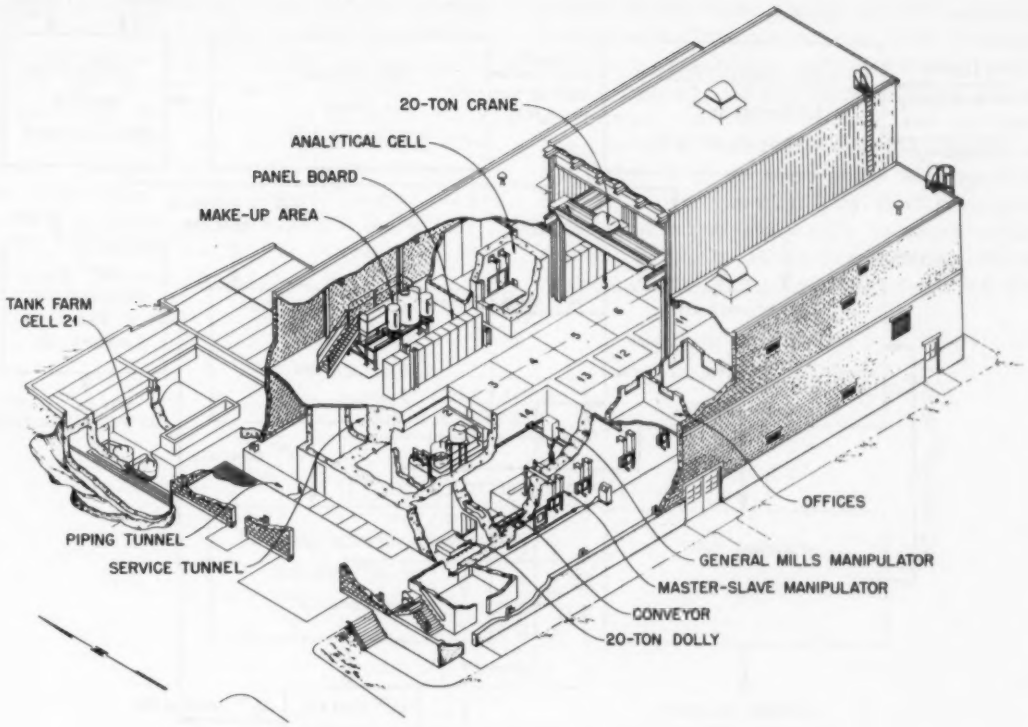


Fig. II-5 Cutaway drawing of Fission Products Development Laboratory.

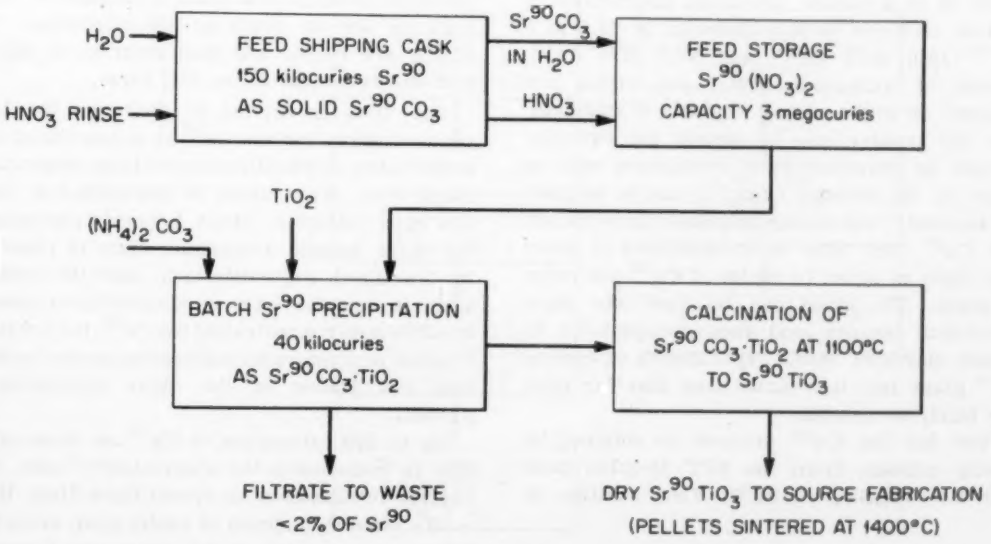


Fig. II-6 Fission Products Development Laboratory Sr^{90} processing.

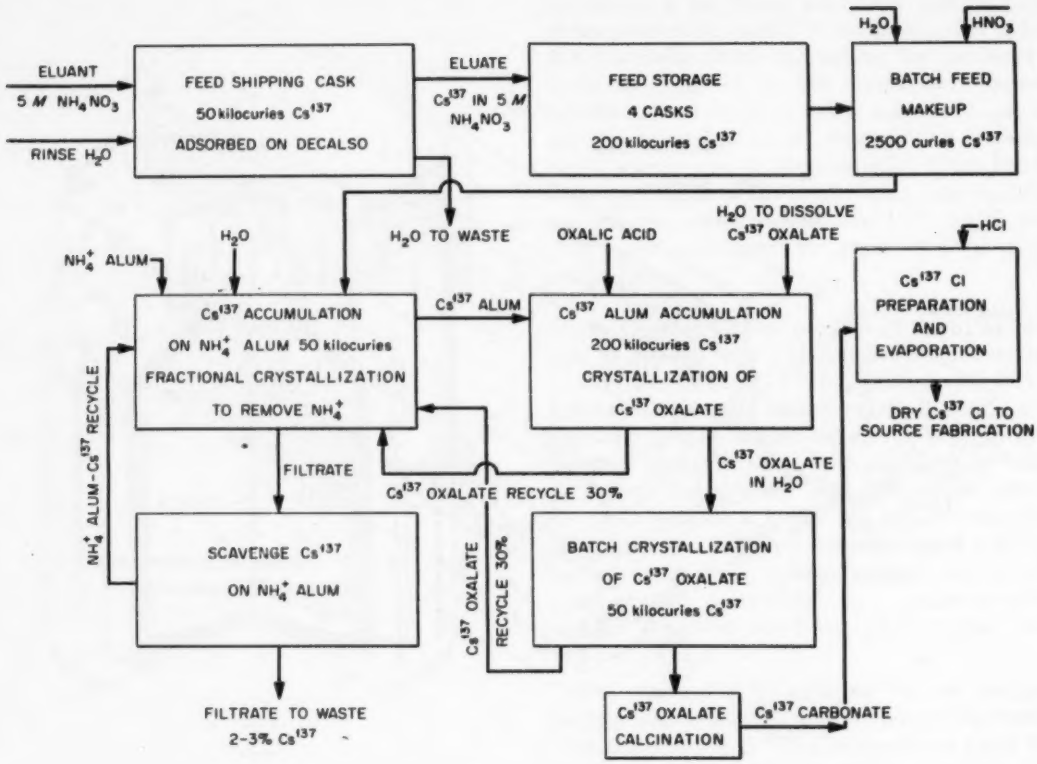


Fig. II-7 Fission Products Development Laboratory Cs^{137} processing.

terial of 26.4 curies. The mass distribution of cesium isotopes in this material is 45.5 at.% Cs^{133}, 16.8 at.% Cs^{135}, and 37.7 at.% Cs^{137}. Traces of aluminum, iron, and nickel are present, as well as small amounts of rubidium. The dry powder can be stored for extended periods in stainless-steel containers with no attack on the storage cans;* it can be weighed and handled by standard manipulator procedures. The Cs^{137} may also be incorporated in glass at a ratio of about 14 curies of Cs^{137} per gram of glass. The glass can be cast into short cylindrical shapes and then encapsulated in welded stainless steel. Experiments on casting Cs^{137} glass into thin slabs have thus far been only fairly successful.

Feed for the Cs^{137} process is obtained by eluting cesium from the STT Decalso beds with 5*M* NH_4NO_3 at 80°C. This solution is stored in underground tanks, which have storage capacity for as much as 500 kilocuries. The STT's are rinsed and then returned to HAPO with the exchanger in the NH_4^+ form.

The feed is diluted to decrease the NH_4^+ concentration, and the Cs^{137} is accumulated by a series of co-crystallizations on large ammonium alum beds. When about 50 kilocuries of Cs^{137} has been collected (about 1 week's operation), the more soluble ammonium alum is removed by fractional crystallization, and the cesium alum is transferred to an accumulation vessel. In addition to concentrating the Cs^{137}, the crystallization process removes fission-product rubidium and traces of the other radioisotopes present.

Up to 200 kilocuries of Cs^{137} as alum solution is collected in the accumulation tanks, and cesium tetraoxalate is crystallized from it in ~70% yield by addition of oxalic acid; since the filtrate is recycled to the mainstream operation, losses are negligible. The cesium tetra-

*See report on cesium sources, page 115.

oxalate is dissolved in water, and the solution is transferred in batches to the final purification cell where cesium oxalate is crystallized again; the filtrate is again recycled to the mainstream. The crystals are calcined in air at 300°C to yield dry, powdered cesium carbonate. This particular form is ideally suited for conversion to any other product form since it is readily soluble in acids.

Dry, powdered $Cs^{137}Cl$ is prepared in 50-kilocurie batches by reacting the $Cs_2^{137}CO_3$ in a water slurry with a slight excess of hydrochloric acid. This operation is done in glassware in a manipulator cell. The $Cs^{137}Cl$ solution is filtered, and the filtrate is evaporated in tantalum equipment to yield the dry salt. The powder is baked with constant stirring at 300°C for 4 hr to remove the last traces of HCl; it is then stored in lots of 10 kilocuries each in stainless-steel cans in water-cooled storage wells. In an earlier method, cesium chloroplatinate was precipitated from the ce-

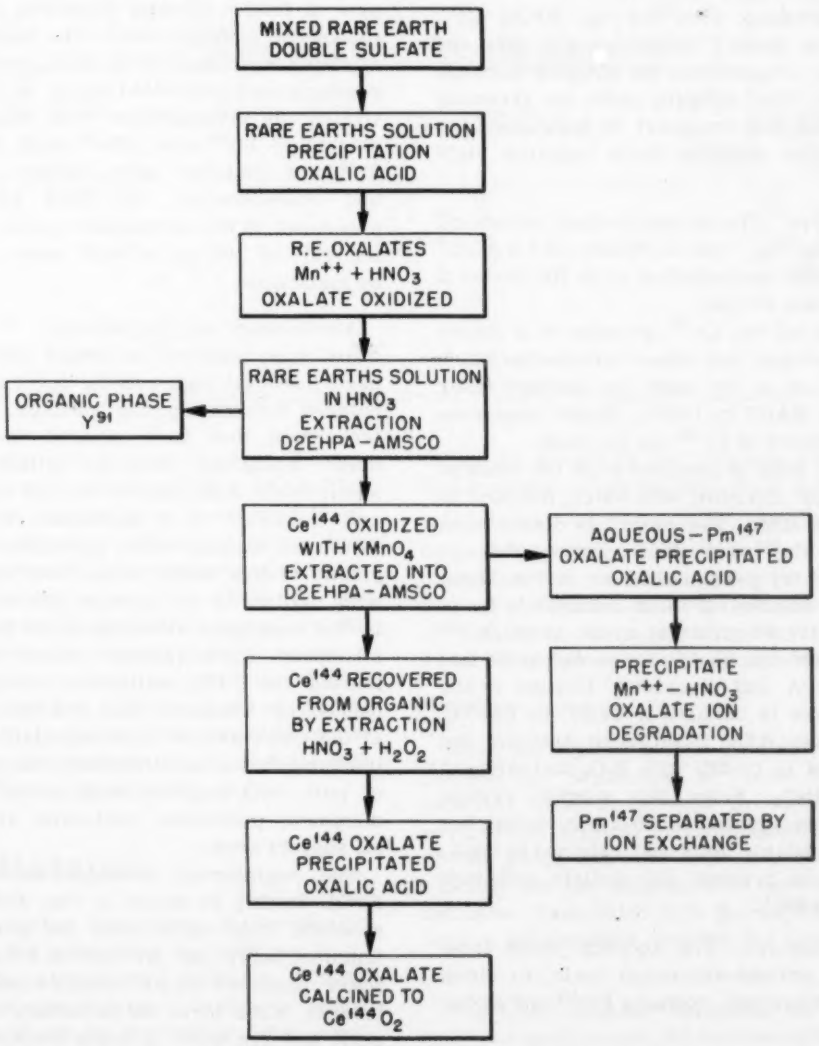

MIXED RARE EARTH DOUBLE SULFATE
RARE EARTHS SOLUTION PRECIPITATION OXALIC ACID
R.E. OXALATES $Mn^{++} + HNO_3$ OXALATE OXIDIZED
RARE EARTHS SOLUTION IN HNO_3 EXTRACTION D2EHPA – AMSCO
ORGANIC PHASE Y^{91}
Ce^{144} OXIDIZED WITH $KMnO_4$ EXTRACTED INTO D2EHPA – AMSCO
AQUEOUS – Pm^{147} OXALATE PRECIPITATED OXALIC ACID
Ce^{144} RECOVERED FROM ORGANIC BY EXTRACTION $HNO_3 + H_2O_2$
PRECIPITATE $Mn^{++} - HNO_3$ OXALATE ION DEGRADATION
Ce^{144} OXALATE PRECIPITATED OXALIC ACID
Pm^{147} SEPARATED BY ION EXCHANGE
Ce^{144} OXALATE CALCINED TO $Ce^{144}O_2$

Fig. II-8 Ce^{144} purification chemistry.

sium alum solution, filtered, washed, and treated with hydrazine to reduce the platinum to the metal. The mixture was filtered, the $Cs^{137}Cl$ was fumed with aqua regia and repeatedly digested with HCl, and the solution was finally evaporated to dryness to yield $Cs^{137}Cl$. Because of the necessity of handling relatively large quantities of HCl and HNO_3 and the time required for evaporating the acids and recovering the platinum, this method has been replaced by the oxalate process.

The Cs^{137} glass is prepared from cesium carbonate by dry-mixing it with silica and divalent oxides and fusing at 1200°C.

The processing rate for the FPDL Cs^{137} operation is about 1 megacurie per year and is dependent primarily on the shipping schedule for the feed. Four shipping casks are presently in use; each will transport 50 kilocuries per shipment. One shipping cycle requires eight weeks.

Cerium-144. The cerium product is sintered pellets of $Ce^{144}O_2$, with a density of 5.5 g/cm^3 and an activity concentration up to 300 curies of Ce^{144} per gram of CeO_2.

The feed to the Ce^{144} process is a double sulfate of cerium and other rare earths which is shipped as a dry solid in shielded filter casks from HAPO to ORNL. Single shipments of 500 kilocuries of Ce^{144} can be made.

The solid cake is removed from the casks at the FPDL by slurrying with water, followed by rinsing with HNO_3. The slurry is dissolved in HNO_3, and Ce^{144} and other rare earths are concentrated by precipitation as the oxalates. A 1*M* HNO_3 solution of these oxalates is fed to the first solvent-extraction cycle, in which Y^{91} and stable yttrium isotopes are extracted into 0.5*M* D2EHPA and discarded. Cerium in the aqueous phase is oxidized to Ce(IV) by $KMnO_4$, extracted into 0.5*M* D2EHPA in Amsco,* and then reduced to Ce(III) with H_2O_2 and stripped with 2*M* HNO_3. From this solution cerium oxalate is precipitated in 20-kilocurie batches in the manipulator cells and calcined to CeO_2. The oxide is pressed into pellets and then sintered at 1400°C.

Promethium-147. The aqueous phase from the second solvent-extraction cycle, in which cerium is recovered, contains Pm^{147} and stable rare earths. These are precipitated as the oxalates and dissolved in HNO_3 to yield a feed solution for the ion-exchange separation of Pm^{147} whenever the latter is needed.

Waste Processing. The FPDL does not currently process raw acid wastes; however, the facility retains the ability to handle such feed streams. Both Redox and Purex type wastes can be processed. The Cs^{137} is removed from the feed stream by co-crystallization on ammonium alum, and the other fission products are then concentrated by a series of precipitations at high *p*H with iron, a normal contaminant of fission-product solutions, as a carrier (magnetite precipitation). The magnetite precipitates are dissolved in HNO_3, and the fission products are separated by a series of controlled *p*H precipitations with NH_3. Fractions containing Ru^{106}-iron, Ce^{144}-rare earths, and Sr^{90} are obtained. After further purification and concentration, the final products are processed in the manipulator cells. A processing rate of 500 gal of acid waste per day can be maintained.

Maintenance and Containment. The FPDL facility was designed to permit direct maintenance, and all high-activity-level cells can be entered through top plug openings. The various techniques that have evolved for removing major equipment such as agitators allow a semiremote maintenance system and limit the cell downtime to a maximum of five days. Personnel making major piping revisions wear plastic airline suits; these suits are equipped with chilled air to increase the working time to the maximum allowable from the radiation standpoint. Contamination control on personnel leaving the FPDL cells has been greatly enhanced by washdown and removal of the suit at the cell entrance. Since standard monitoring instrumentation can detect only large quantities of pure beta emitters such as Sr^{90} or Pm^{147}, maximum preventive measures are taken in all suspect areas.

The containment arrangement used in the FPDL facility is shown in Fig. II-9. The 4-ft concrete cell walls form the primary containment layer and are held at 1.5 in. negative water pressure under normal conditions. The building walls form the secondary containment shell and are under a slight negative pressure during normal operations. All air entering the building is filtered through CWS type filters;

*Amsco-125, boiling-point range, 342 to 404°F.

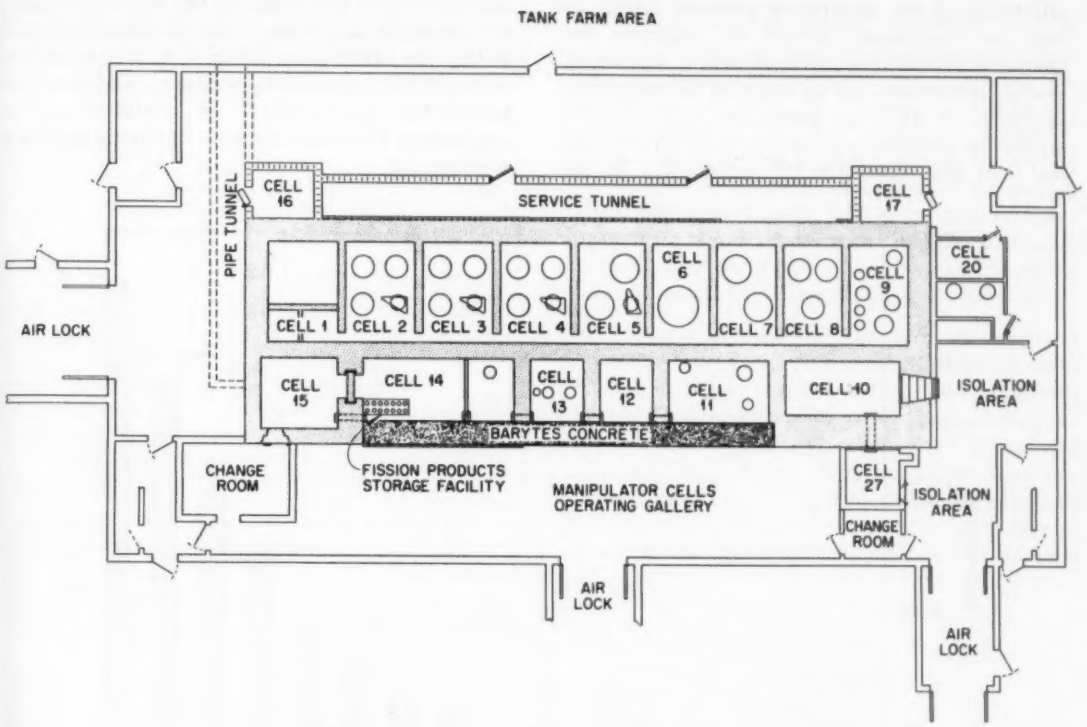


Fig. II-9 Containment arrangement in the Fission Products Development Laboratory.

air leaving the cells is also filtered through CWS type filters before entering the ORNL cell-ventilation system. In an emergency, high-velocity air monitors signal an alarm that actuates the automatic containment system: fans shut down, inlet air dampers close, an additional cell-ventilation damper opens, and the building pressure is reduced to 0.5 in. negative water pressure in less than 10 sec. Under these conditions the cells are held at 0.8 in. negative water pressure with respect to the building, and the building is at -0.3 in. H_2O with respect to the atmosphere.

Stable Isotopes

Although popular conception of the atomic energy program associates the isotopes business with the glamorous "twinkling atoms," the increasing importance of separated stable isotopes should not be overlooked; these silent partners play a rather important role in the overall applications program. Figure II-10 shows the growth of the program as indicated by the number of annual shipments since 1946.

Most stable isotopes are separated in electromagnetic devices called "calutrons,"* or to a limited extent by other methods, including thermal diffusion, centrifugation, and countercurrent fused-salt migration. By far the most versatile of these involves the calutron.

History

Since the replacement of the electromagnetic process for separating U^{235} from U^{238} by the gaseous diffusion method in 1946, a small part of the original electromagnetic facilities at Oak Ridge has been kept in operation specifically for the separation of enriched stable isotopes. Associated with the separations has been a development program for improving the

*The term "calutron" designates the production type mass spectrometer, the term having been coined at the time of its development by E. O. Lawrence at the University of California.

efficiency of the separation process itself, as well as continuous efforts to improve the chemical procedures involved both in preparing charge materials and in refining the separated isotopes. In 1958 an expansion program was started to reactivate a number of beta calutrons that had been used for U^{235} production during

and receivers for retaining the separated ions, all operating in a vacuum. The notable difference is that the calutron is designed to collect usable amounts of separated isotopes, whereas the laboratory spectrometer is designed as an instrument for mass analysis of minute amounts of material.

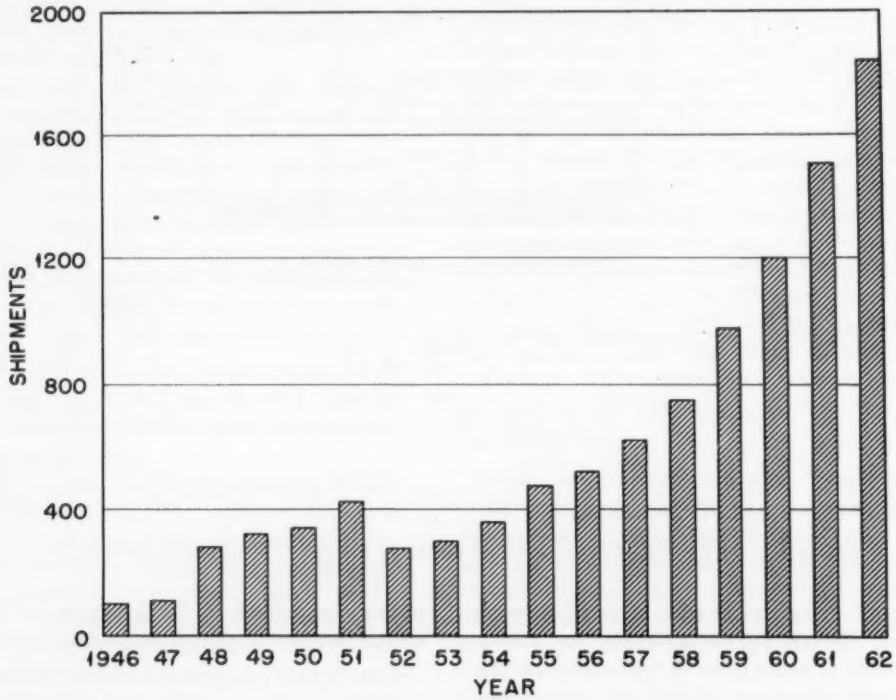


Fig. II-10 Shipments of electromagnetically separated stable isotopes (1946–1962).

World War II. At present 28 units, including the original four that have been in continuous use since 1946, are assigned to stable isotope separations; 8 (doubly contained) are allocated to processing plutonium, thorium, uranium, and other radioactive materials; and 6 are used for research and development activities. The separation capacity has been increased markedly; as a result of the larger scale of operation, the separated isotopes are, in general, being produced more cheaply.

The basic principle behind the electromagnetic separation of isotopes is identical to that of an analytical mass spectrometer. Both depend on high potentials for acceleration, magnetic fields for separating isotopic ion species, ion sources for ionizing the charge materials,

Figure II-11 is a schematic diagram illustrating the electromagnetic separation process. Positively charged ions are accelerated by voltages of up to 40 kv. As these rapidly moving, singly charged ions move through the magnetic field existing between the source and receiver, they are deflected, the amount of deflection for a particular field strength and potential depending only on the masses of the ions. Thus isotopes of a particular element traverse slightly different paths and can be intercepted in such a way as to accomplish a separation.

The element to be processed (or a suitable compound) is usually evaporated by resistance heating of a "charge bottle," and the vapor is subjected to an ionizing beam of electrons,

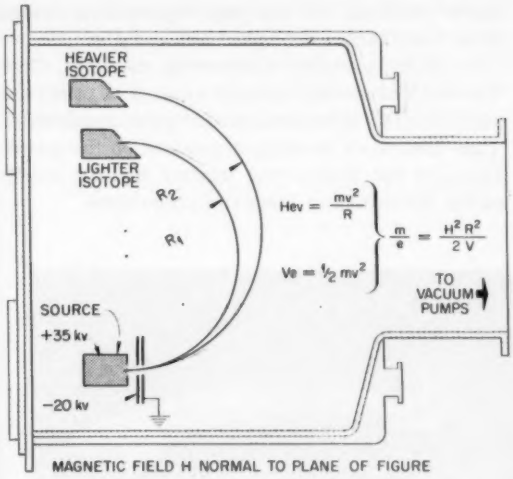


Fig. II-11 Illustration of the process of the electromagnetic separation of isotopes.

Various receiver-pocket materials may be used, depending on the element being processed and the chemistry involved in recovering the separated isotopes. For example, silver receiver pockets are used to collect mercury, which amalgamates with the silver, improving the retention. Graphite is used most frequently since the carbon can be burned in oxygen, leaving the residual oxide of the collected isotope. Copper is used in cases where simple leaching with dilute acid is feasible, as with calcium.

Unfortunately not all the ions that enter a receiver pocket are retained; some of them bounce out again before "sitting down." Others, by physical erosion, dislodge some of the material already collected. As a result the amount of material actually retained in the receiver pocket may vary from 10% for some antimony and magnesium runs to >99% for some platinum runs. From 1946 to 1956, only 56% of the ions

Table II-2 TYPICAL CHARGE MATERIALS

Element	Charge material	Element	Charge material
Antimony	Sb; Sb_2O_3; Sb_2S_3; SbI_3	Nickel	$NiCl_2$; $Ni(CO)_4$
Barium	Ba; $BaCl_2$; $BaBr_2$	Platinum	Pt†
Bromine	$SrBr_2$*	Potassium	KI; K; KBr; KCl
Cadmium	$CdCl_2$; $CdBr_2$; CdI_2; Cd	Ruthenium	Ru†
Calcium	Ca; $CaCl_2$; CaI_2	Silicon	$SiCl_4$
Cerium	$CeCl_3$	Silver	AgCl; Ag; AgBr; AgI
Chromium	$CrCl_3$	Strontium	Sr; $SrBr_2$
Copper	$CuCl_2$; Cu_2Cl_2	Tellurium	TeO_2; Te; $TeBr_4$
Europium	$EuCl_3$	Tin	$SnCl_2$; $SnCl_4$
Iron	$FeCl_2$; $FeCl_3$	Titanium	$TiCl_4$
Lead	$PbCl_2$; PbI_2; $(C_2H_5)_4Pb$	Tungsten	WCl_6; WBr_6; WF_6; WO_3
Magnesium	Mg; $MgCl_2$; $MgBr_2$; MgI_2	Zinc	Zn
Mercury	$HgCl_2$; HgO	Zirconium	$ZrCl_4$

*Strontium is also collected.
†Electron bombardment source.

which dislodges electrons from the atoms, leaving them positively charged. These ions are now ready for acceleration into the magnetic field. Typical charge materials are listed in Table II-2.

Occasionally, as in the case of the platinum metals, extremely high temperatures are required for evaporation, and the heat is supplied by electron bombardment of the element. Figure II-12 is a drawing of such a bombardment-heated source.

Once the individual isotope beams have been separated by the magnetic field, they enter the receiver "pockets" (Fig. II-13).

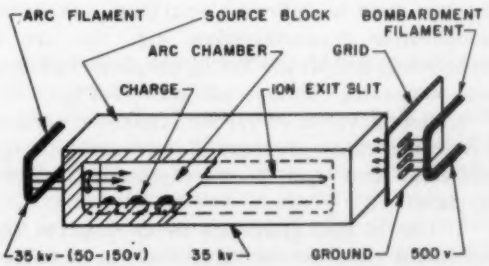


Fig. II-12 Bombardment-heated source used for charge materials requiring extremely high temperatures for evaporation.

entering receiver pockets were retained as separated isotopes in the ORNL separators.

Table II-3 lists the isotopes of several elements and includes typical data to show operating parameters and results of the separations.

Table II-4 shows the cumulative results of modifications in operating techniques as exemplified by higher grade products. Some of the factors producing these improvements are:

1. Better feed materials: higher purities, e.g., removal of the 1% strontium in ordinary barium feed to decrease sparking; and better evaporation characteristics, e.g., the use of completely anhydrous $FeCl_2$, prepared by fusing and outgassing commercial-grade $FeCl_2$.

2. Refrigeration of vacuum-chamber walls to lower the vapor pressure of unresolved charge material that has been evaporated into the system.

3. Use of dual filaments in the source. The operating time between shutdowns is increased by having a second filament ready to operate when the first one burns out.

4. Introduction of an electron bombardment source to increase the vaporization (and hence beam current) of low-vapor-pressure charge materials such as the platinum metals.

5. More precise monitoring methods. Continuous and careful manual control of operating parameters is essential to good separation. This control is directly dependent on the sensitivity of the monitoring system and the ability of the operators to discern fluctuations.

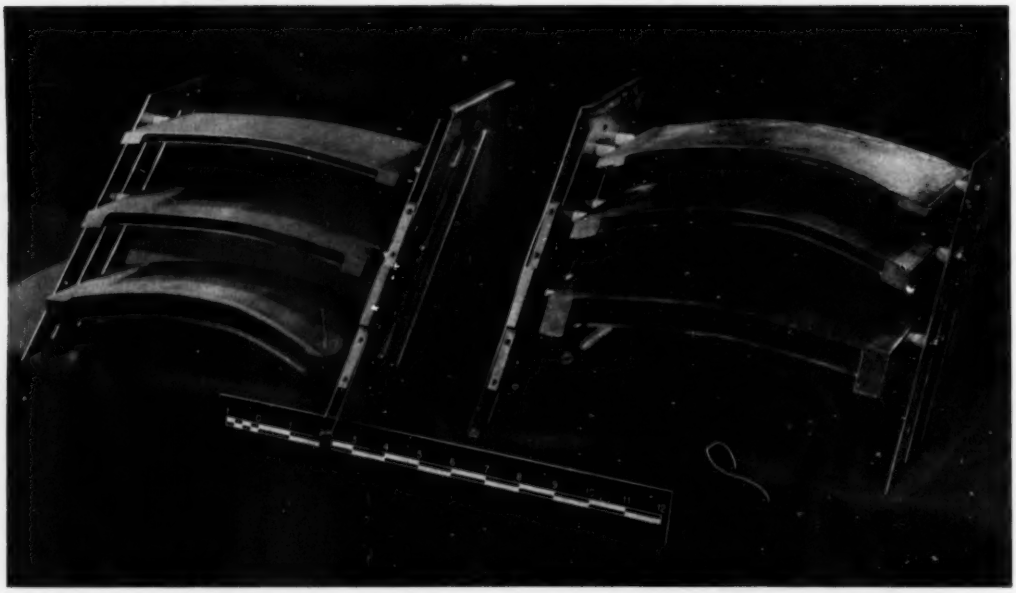


Fig. II-13 Typical receiver pockets for the collection of isotopes. The spacing between the slots depends upon the isotopes being collected.

6. Improved receiver-pocket design; improved geometry and increased cooling; and sputtering receiver materials onto cold surfaces, thereby improving retention and reducing contamination.

As knowledge applicable to recovery problems has accumulated through experimentation, the retention of isotopic material has gradually increased. Over the past two-year period, retention has averaged 80%. The net gain in retention cannot be attributed to any single factor but represents the combined effects of engineering improvements incorporated to minimize the losses associated with the sputtering of receiver-pocket walls and the low retention of isotopes on surfaces struck by ions.

In some investigations into retention problems, ion currents associated with the more

abundant isotopes and relatively high current densities have been utilized. Several studies have recently been made with a less abundant isotope, Ca^{46} (natural abundance, 0.0033%). Investigations were made with the Ca^{46} ion beam at a current density of only 0.22 $\mu a/cm^2$ on graphite, copper, and crystalline copper having the 1 1 0 and 1 1 2 crystal faces set perpendicular to the incident ion beam. The amount of Ca^{46} striking the target surface was monitored, and quantities and purities of Ca^{46} recovered from the target area and from the pocket area were determined. In Fig. II-14 the quantity and purity of Ca^{46} recovered from the surfaces of targets made of normal copper, purified graphite, and a 1 1 0 copper crystal are compared. The purity of samples recovered from each surface is essentially constant, but there appears to be more Ca^{46} retained per unit area on graphite than on either copper or a 1 1 0 copper crystal. In Fig. II-15 the isotopic purity and the ratio of wanted to unwanted isotopes are compared to show equality of target materials from this standpoint.

In Fig. II-16 the isotopic purity of Ca^{46} recovered from the target area is compared with that recovered from the pocket area. Extreme differences in purity are noted, and in every determination the Ca^{46} recovered from the area struck by the beam is of higher purity than that recovered from the pocket area. In Fig. II-17, where the ratios of wanted to unwanted isotopes in the recovered samples are compared, the trend noted in the previous

Table II-3 TYPICAL OPERATING PARAMETERS AND RESULTS OF ISOTOPE SEPARATIONS

Isotope	Average beam current, ma	Receiver pocket	Natural isotope abundance,* %	Enriched isotope abundance, %	Rate of product recovery, mg/hr
Copper	100	Graphite			30
Cu^{63}			69.0	99.9	
Cu^{65}			31.0	99.6	
Iron	100	Graphite or copper			100
Fe^{54}			5.9	93.8	
Fe^{56}			91.6	99.8	
Fe^{57}			2.2	85.0	
Fe^{58}			0.33	75.0	
Platinum	9	Graphite			25
Pt^{190}			0.012	0.8	
Pt^{192}			0.78	13.8	
Pt^{194}			32.8	65.0	
Pt^{195}			33.7	60.1	
Pt^{196}			25.4	65.9	
Pt^{198}			7.2	60.9	
Potassium	30	Copper			10
K^{39}			93.2	99.9	
K^{40}			0.012	7.7	
K^{41}			6.7	99.0	
Silver	45	Copper			50
Ag^{107}			51.4	99.0	
Ag^{109}			48.6	99.2	
Zinc	40	Copper			40
Zn^{64}			48.9	99.8	
Zn^{66}			27.8	98.8	
Zn^{67}			4.1	92.4	
Zn^{68}			18.6	98.5	
Zn^{70}			0.63	78.3	

*An isotopic abundance is the atom percent of that isotope present in the total mixture of isotopes.

Table II-4 IMPROVEMENTS IN ISOTOPIC PURITIES

Isotope	Natural abundance, %	Enriched abundance, %	
		Original	Improved
Ag^{107}	51.4	88.9 (1946)	98.8 (1955)
Ag^{109}	48.6	95.8 (1946)	99.9 (1955)
Eu^{151}	47.8	78.7 (1955)	97.8 (1956)
Eu^{153}	52.2	86.4 (1955)	99.9 (1956)
Fe^{54}	5.9	42.8 (1946)	92.7 (1954)
Fe^{56}	91.6	98.7 (1946)	99.7 (1954)
Fe^{57}	2.2	56.1 (1946)	72.6 (1954)
Fe^{58}	0.33	22.0 (1946)	81.5 (1954)
Cu^{63}	69.0	97.0 (1945)	99.9 (1960)
Cu^{65}	31.0	93.1 (1945)	99.6 (1960)

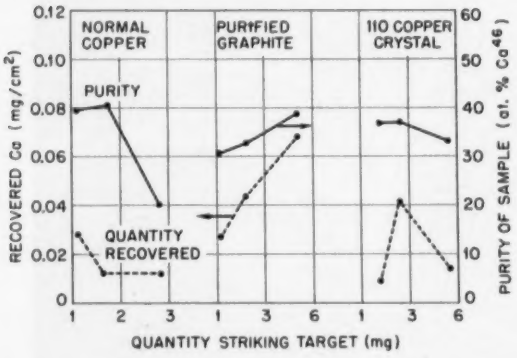


Fig. II-14 Comparison of purity and quantity of Ca^{46} recovered from various target surfaces.

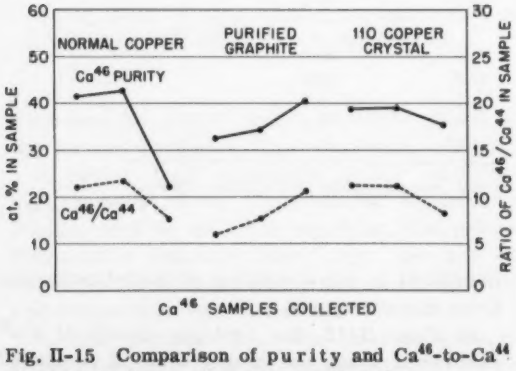


Fig. II-15 Comparison of purity and Ca^{46}-to-Ca^{44} ratio obtained on various target materials.

data is again supported; namely, a better-quality sample is obtained by selectively recovering the material from the area struck by the ion beam.

Although there are insufficient data to prove that oriented copper crystals have an advantage

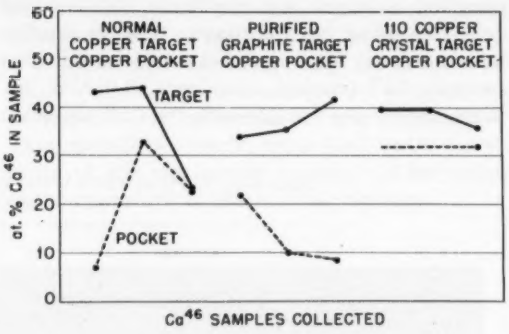


Fig. II-16 Comparison of isotopic purity of Ca^{46} recovered from target and from pocket surface.

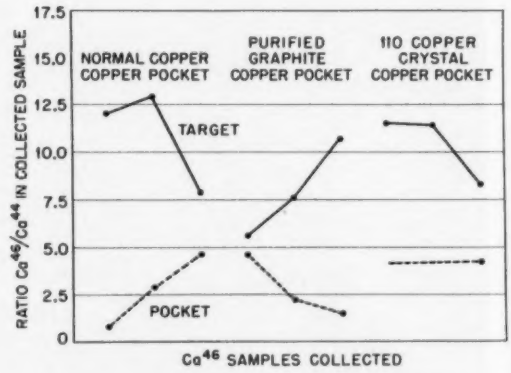


Fig. II-17 Comparison of Ca^{46}-to-Ca^{44} ratio in samples recovered from pocket surface.

over other collector materials, scattered evidence indicates that they offer some, even if slight, improvement. The actual processes producing such improvement are not yet understood, but they are sufficiently important to warrant further efforts to establish definite conclusions.

A special collector has been designed and is currently being used in the calcium separation to determine saturation values for Ca^{46} on various copper crystals. The receiver is constructed so that four different targets can be mounted on a rotating holder. The holder rotates 90°, holds for a few minutes, and then repeats. Each target is thus in position so that the beam strikes it normal to its surface for 6 min of each rotation. Since operating conditions vary from run to run, this technique provides comparative data that are independent

Table II-5 SECOND-PASS SEPARATIONS

Isotope	Natural abundance, %	First-pass enrichment, %	Second-pass enrichment, %	Abundance of undesirable isotopes, %	
				After 1st pass	After 2d pass
Fe^{58}	91.6	99.7	99.987	0.3	0.013
Cr^{52}	83.7	99.5	99.97	0.5	0.030
Li^{6}	7.5	95.3	99.999	4.7	0.001

Table II-6 TYPICAL PRODUCT FORMS

Element	Product form	Element	Product form
Antimony	Sb	Nickel	NiO; Ni
Barium	$Ba(NO_3)_2$; $BaCO_3$	Platinum	Pt
Bromine	AgBr; NH_4Br	Potassium	KCl; K_2SO_4
Cadmium	CdO; Cd	Ruthenium	Ru
Calcium	$CaCO_3$	Silicon	SiO_2
Cerium	CeO_2	Silver	Ag; AgCl
Chromium	Cr_2O_3	Strontium	$SrCO_3$; $Sr(NO_3)_2$
Copper	CuO; Cu	Tellurium	Te
Europium	Eu_2O_3	Tin	SnO_2; Sn
Iron	Fe_2O_3; Fe	Titanium	TiO_2
Lead	$PbCO_3$; PbO; Pb	Tungsten	WO_3
Magnesium	MgO	Zinc	ZnO
Mercury	$Hg(NO_3)_2$; Hg	Zirconium	ZrO_2

Table II-7 ELEMENTS SEPARATED DURING THE YEARS 1946 TO 1962

1946	Br*, Cd*, Ca*, C*, Cr*, Cu*, In*, Fe*, Pb*, Li*, Mg*, Mo*, Ni*, K*, Sc*, Si*, Ag*, Sr*, Sn*, Zn*, Zr*
1947	Sb*, B*, Cd, Ca, Cu, Ge*, In, Fe, Pb, Li, Mg, Hg*, Mo, Ni, K, Se, Si, Sr, Te*, Tl*, Sn, W*, Zn, Zr
1948	Ba*, Ca, Ce, Cl*, Cu, In, Fe, Pb, Li, Hg, Ni, O, K, Re*, Se, S*, Sn, W, Zr
1949	B, Ce, Cu, Fe, La*, Li, Mg, Hg, K, Se, Si, Ag, Te, Sn, Zr
1950	Br, Ca, C, Cr, Ga*, Hf*, In, Fe, La, Pb, Li, Nd*, K, Rb*, Sm*, Sr, S, Ti, W, V*, Zn
1951	Ca, C, Ce, Cu, Fe, Li, Ni, Sm, Se, Sr, Te, V
1952	Ba, B, Cl, Ge, Pb, Li, Mo, Nd, Si, S, Sn, Ti
1953	B, Gd*, Hf, Ir*, La, Li, Nd, Pd*, Pt*, K, Ru*, Sm
1954	Ca, Cr, Cu, Fe, Ni, Rb, Ta*, Zr
1955	Ba, Br, Cr, Li, Mg, Mo, N*, Sm, Ag, Sr, S, Te, Ti, W
1956	Sb, B, Cd, Ca, Cl, Cu, Dy*, Eu*, Ge, Pb, Li, Pd, Pt, Re, Rb, Ru, Si, Tl, Sn, Zn
1957	Fe, Ir, Ti, Cr, C, Er*, Se
1958	V, Yb*, Lu*, Ca, K, Ag, Cu, Hf, C, Se, Mg, Gd
1959	Rb, Bi, Tl, Hg, Sr, Nd, Ni, Cu, Ge, Sm, Zn, In, Ba
1960	Pb, Cr, Os*, Ta, Cu, Fe, Ga, Ni, Cd, Zn, K, Cl, Si, Br
1961	Yb, Zr, Cl, Mg, Sn, Sm, Ni, W, Gd, Lu, Ti, Ta, Hf, Ca, Mo, La, Re
1962	Nd, Si, Ca, Sr, Er, Fe, Te, Cu, Ta, Yb, Cd

*Separated for the first time.

NOTE: In a number of instances, an element was separated more than once in the same year.

of operating conditions for different receiver materials.

Table II-5 lists isotopes that have undergone "second-pass" separations and shows original, intermediate, and final enrichments.

The procedures for chemical recovery usually involve rather straightforward inorganic techniques, although they must be modified because of the high value of the enriched material. Lead, for example, is recovered by leaching copper receiver pockets with HNO_3, electrodepositing the lead at the anode as PbO_2, dissolving the PbO_2 in hot HNO_3 and H_2O_2, and finally crystallizing high-purity $Pb(NO_3)_2$ from 85% HNO_3. Copper is recovered by igniting the graphite receiver pockets in oxygen, dissolving the ash in HNO_3, and electrolyzing the copper. This copper is then redissolved in HNO_3, precipitated with cupferron, and ignited to CuO.

Silicon is collected in graphite pockets. This is burned in oxygen, and the SiO_2 residue is fused with Na_2CO_3. The melt is then dissolved in water, and SiO_2 is precipitated by acidification and digestion with $HClO_4$.

The chemical recovery of rare earths often involves ion-exchange purifications to remove adjacent rare-earth impurities. Even then, high-purity products are difficult to obtain.

Table II-6 lists typical product forms for 26 elements. Table II-7 shows the elements separated in the years 1946 to 1962.

One of the most significant contributions of this phase of the isotopes program is the use of enriched stable-isotope targets for the production of radioisotopes. Shorter irradiation times, higher specific activities, and freedom from undesirable extraneous activities are characteristic of radioisotopes prepared by bombardment of separated stable nuclides. The decreased extraneous activity usually results from both increased concentration of desired target nuclide and decreased concentration of undesired target nuclide. Table II-8 lists a few of the enriched nuclides used for radioisotope production, along with information to show the advantages of using these targets rather than normal material. The same advantages apply with respect to cyclotron-produced radioisotopes.

Table II-8 ENRICHED STABLE ISOTOPES USED IN RADIOISOTOPE PRODUCTION

Radioisotope	Target nuclide	Percent abundance		Advantages	
		Natural	Enriched	A*	B†
Cd^{109}	Cd^{108}	0.88	70	80	Cd^{113}: 400 Cd^{115}: 550
Ca^{45}	Ca^{44}	2.1	98	47	
Ca^{47}	Ca^{46}	0.003	30	10,000	Ca^{45}: 3,000
Cr^{51}	Cr^{50}	4.3	95	23	
Fe^{55}	Fe^{54}	5.8	98	17	Fe^{59}: 18,000
Fe^{59}	Fe^{58}	0.31	76	250	Fe^{55}: 2,500
Ni^{63}	Ni^{62}	3.7	98	25	Ni^{59}: 2,500
Sr^{85}	Sr^{84}	0.55	54	100	Sr^{89}: 300

*A is the approximate factor by which specific activity can be increased for a given irradiation time or by which irradiation time can be decreased for a given specific activity.

†B is the approximate factor by which the concentration of an extraneous (unwanted) radionuclide is reduced relative to the desired nuclide.

These advantages are illustrated nicely by Ca^{47}. When prepared from the 0.003% Ca^{46} in normal calcium, the 4.5-day isotope is completely overshadowed (by a ratio of about 25:1) by the 164-day Ca^{45} produced by irradiation of the 2.1% Ca^{44} also present. However, if the Ca^{46} is enriched to, say, 30%, the Ca^{44}-to-Ca^{46} ratio is reduced to about 1:5, and the resulting Ca^{45}-to-Ca^{47} ratio becomes about 1:125; thus a useless product that is 96% undesirable Ca^{45} can become a very useful product that is 99% Ca^{47}.

Table II-9 shows some typical uses for stable nuclides.[5] A recent bibliography[6] covering these and other applications is included in USAEC Report ORNL-3266.

During the first 10 years of the electromagnetic separations program, nearly one fourth of the isotopes were used to determine energy levels and another one fourth to measure various neutron cross sections.[8] Today much of the effort is involved in the various cross-section studies.[9,10] However, very interesting work continues on energy levels,[11] decay schemes,[12] and half-lives,[13] and on such other topics as hyperfine structure,[14] the measurement of bulk properties,[15] and Mössbauer studies.[16,17]

The use of enriched stable isotopes in isotope dilution as an analytical tool has good potential.[18] Their use in tracer work involving eventual activation analysis is also a powerful tool.[19] A unique use of B^{10} consists of introducing a soluble compound into the bloodstream of a patient with a brain tumor, allowing the isotope to concentrate in the tumor, and then irradiating the B^{10} with neutrons to provide an on-the-spot treatment of the tumor.

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Table II-9 TYPICAL USES FOR ENRICHED STABLE ISOTOPES*

Target materials for preparing high-specific-activity radioisotopes in reactors and in accelerators. Enriched targets not only increase the number of activations per unit time of irradiation but also reduce side activities introduced by other isotopes. Calcium-44, Cr^{50}, Cr^{54}, Fe^{54}, and Fe^{58} are examples of enriched isotopes used in producing radioisotopes.

Studying the isotope effects on bulk properties of elements. Specific heat, density, and the like have been studied for Li^6 and Li^7; the isotopes of lead, mercury, and tin have been investigated in connection with superconductivity; Ni^{58} and Ni^{60} have been used to study the isotope effect on linear expansion.

Physicochemical studies, such as reaction kinetics and self-diffusion.

Starting materials in assigning masses to artificially produced radioactive isotopes. The results of a mass spectrometric analysis of irradiated material can be positively correlated to an activity only when the target material is mononuclidic. More than 100 such mass confirmations have been made in this way.

Isotope-dilution analyses. Traces of enriched K^{39}, K^{41}, and N^{15} have been used in fertilizer uptake studies; K^{41} in geologic age determinations; V^{50}, Fe^{54}, and Ni^{62} in estimating trace impurities in crude oils.

Studying naturally occurring, low-abundance radioisotopes such as K^{40}, Sn^{124}, and Sm^{147}. It is much easier to evaluate the properties of these isotopes using enriched material than it is using the normal element. This is particularly true in the case of the double-beta-decay studies of K^{40}.

Preparing calibration standards for mass spectrometers. "Blends" of various concentrations of essentially pure nuclides provide reference standards. An example is mixtures of enriched Li^6 and Li^7 to give a complete range of relative abundances.

Preparing radioisotopes for studies of decay schemes and characteristic radiations. Numerous highly enriched nuclides have been activated, and the investigators have thereby been spared the tremendous confusion that results from trying to unscramble the decay schemes of several species that are disintegrating simultaneously.

Medical use. Gallium-71 has been used in bone-cancer studies; K^{40} has been used in tissue studies of malignant lymphoma.

Sources of monochromatic light for optical calibration. Wavelength standards are preferable when uninfluenced by any isotope effect due to isotopic impurities. Cadmium-114, Hg^{198}, and Hg^{202} have been used as standards. The orange-red wavelength of Kr^{86} has been made the basis for the official definition of the meter.[7]

Optical studies of isotope shift, hyperfine structure, and nuclear spin. Hundreds of publications on these subjects exist.

Measuring neutron absorption, scattering, and total cross section. This probably involves the most concentrated efforts at the present time; enriched samples of almost all the stable isotopes are used.

"Pushing back" the limits of natural abundance of certain isotopes. The lower limits of possible abundance of a particular species can be ascertained with more assurance by analyzing material collected in the position for a "nonexistent" isotope. This has been done for Yb^{167}, Er^{172}, and Pb^{210}. The technique has also been used for Ta^{180} to help assign an actual value of abundance to an isotope previously thought to be nonexistent.

Spectrochemical standards. Spectrochemically pure Ca^{40} is recovered from the electromagnetic separation of calcium isotopes.

*With minor changes, reprinted with permission from *Chem. Eng. News*, 37(5): 64 (1959).

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Section

III

Isotopes and Radiation Technology

Isotope Technology Development

The general principles of isotope applications were reviewed in Sec. I; in the Tokyo speech by Fowler and Aebersold, isotope technology was divided into the following fields: neutron activation analysis, radiometric analysis, process control with low-level tracers, radiation absorptiometry, ionization technology, nondestructive testing by radiography, and isotope measurement systems. In this issue some current work on the use of isotopes in a new analytical chemical method that has been named the "radiorelease" technique and in activation analysis is reviewed. In succeeding issues other of these fields will be covered.

Analysis

Process control by chemical or physical analysis is a very important and frequently a costly part of a manufacturing process. Much work is being done under the sponsorship of the Division of Isotopes Development, U. S. Atomic Energy Commission (USAEC), to develop ways for using radioisotopes to simplify and improve analytical methods both in industrial process control and in the determination of contaminants in our environment. New developments in the use of radioisotopes in analysis are reviewed biennially in *Analytical Chemistry*, and the number of references in these reviews reflects the large amount of activity in this field. The most recent review by Leddicotte,[1] in 1962, and three earlier reviews by Meinke[2] contain over 4000 references; in the two most recent compilations, coverage is limited to selected articles rather than to all papers in the field.

The use of radioisotopes in analysis frequently offers some or all of the following advantages:

1. More sensitive because of the possibility of detecting very low radiation levels
2. Faster because of the ease of detection of radioisotopes
3. More easily instrumented and automated, which permits easy feedback for process control
4. More specific since radioisotopes of chemically similar elements may have entirely different properties
5. Often nondestructive because the counting can frequently be done without separation

These advantages make radioisotopes potentially very useful in process control and often result in analyses being moved out of the laboratory and into the process.

In this issue several novel uses of the radiorelease technique are discussed, and some of the newer developments in neutron activation analysis are surveyed.

Radio-Release Techniques

In the radio-release method of analysis, a nonradioactive substance is determined indirectly by the quantitative release of a radioactive material from a reagent with which it is treated. For example, ozone oxidizes hydroquinone in which Kr^{85} has been trapped:

$$[C_6H_4(OH)_2]_3Kr^{85}(s) + O_3(g) \rightarrow 3C_6H_4O_2(s) + 3H_2O(l) + Kr^{85}(g)$$

Such "compounds," in which one compound or element is trapped in the crystal lattice of another, are known as "clathrate" or "cage" compounds. The Kr^{85}, which is released quantitatively, is easily measured by standard counting techniques, and from this measurement the ozone can be calculated. Since the analysis technique does not introduce radioactivity into the process, it is particularly attractive for process control.

Because of its relatively long half-life (10.6 years), its chemical and biological inertness, and its moderate-energy beta (0.67 Mev) and absence of gamma energy, Kr^{85} is an ideal isotope for such purposes.

Chleck and coworkers at Tracerlab, Inc., have developed methods for making Kr^{85} clathrates suitable for use as reagents[3] and, using these clathrates, have developed analyzers for small amounts of ozone[4] and sulfur dioxide.[3] Workers at Parametrics, Inc., have developed methods for making radio-release reagents by trapping Kr^{85} in metals by ion bombardment or by high temperature and pressure diffusion.[5] Rotariu and coworkers at Booz-Allen Applied Research, Inc., applied Kr^{85} clathrates to the analysis of liquids,[6] and Richter and Gillespie at Research Triangle Institute have developed a radio-release method for determining small amounts of dissolved oxygen in water.[7]

Preparation and Properties of Kr^{85} Clathrates. In 1950 Powell published procedures for making hydroquinone clathrates of argon, krypton, and xenon.[8] An almost saturated aqueous solution of hydroquinone was heated to 90°C and then slowly cooled in contact with the inert gas at a pressure of about 40 atm. The product was a stable crystalline substance in which molecules of the rare gas were trapped in the hydroquinone crystal. Many compounds of this type can be prepared by similar techniques, as discussed in a recent book.[9]

Chleck, Ziegler, and coworkers,[3,10,11] to avoid the added manipulative steps of the solution method when working with the radioactive gas, devised the following procedure for crystallizing the Kr^{85} clathrate from the melt:

About 1 g of hydroquinone is put in a pressure bomb, atmospheric gases are removed, and Kr^{85} is introduced under pressure. The bomb is heated to 185°C to melt the hydroquinone and held at this temperature for 2 hr to allow the Kr^{85} to saturate the melt. The system is then cooled slowly to room temperature, the bomb is opened, and the sample is removed. With pressures up to 60 atm and cooling for as long as 72 hr, the greatest clathrate trapping efficiency* was 28.3%. This would provide 25.8 equivalent atmospheres of Kr^{85} at 15°C. With 20 atm pressure and 17 hr cooling, the efficiency was 17%. At 10 atm pressure and 4 hr cooling, efficiency was poor, but some clathrate formed.

The clathrates thus formed were a fused hard mass, with activities of 30 μc/g, 15 mc/g, and 1.5 curies/g, depending on the specific activity of the original krypton and the efficiency of the process. The leakage rate of Kr^{85} from the strongest sample (1.5 curies/g) was about 3 μc/(g)(day). This rate (10 times that of the 15 mc/g sample on a per unit activity basis but less than that of the weakest sample) is low enough that these clathrates can be kept in a closed room for weeks without exceeding the maximum allowable concentration. The samples are stable at room temperature and are resistant to radiation breakdown. Wilson and Hughes[12] at Harwell, using Powell's solution procedure, obtained a clathrate efficiency of 80 to 90%. However, the compounds were not stable, with 10% of the Kr^{85} being lost in the first month and another 5 to 10% in the next two months. They attributed the instability of their compounds to the effect of the beta radiation from the Kr^{85} on the water trapped in the compound.

Mock, Myers, and Trabant at Purdue[13] studied crystallization from a number of different solvents, crystallization from melts, and sublimation. They found that crystallization from a solvent gave the best efficiency, and from a melt, the poorest. They also tried a number of other crystals, but none was as good as hydroquinone. Nonradioactive samples of these clathrates exposed to 10^8 rads in the Purdue University gamma facility showed no weight change.[14]

Kryptonates as Universal Radioactive Indicators. Another type of solid compound of krypton which should have wide application as a radio-release reagent has been developed by Chleck and associates at Parametrics.[5] These workers incorporated krypton, containing 5% Kr^{85}, in solid materials both by bombardment with ionized krypton and by diffusion under high temperature and pressure. The ionization procedure is now used only for substances that cannot withstand the pressures or temperatures required in the more efficient diffusion process.

Typical conditions for the diffusion saturation of a substance with krypton are tempera-

*The efficiency of a clathrate is defined as the amount of gas trapped compared to the theoretical amount possible.

tures around 650°C and pressures up to 5000 psi for from several hours to three days. The possible saturation is a function of the structure of the compound, with substances having numerous interstices of the proper dimensions to contain the krypton atom becoming the most highly saturated. Pyrolytic graphite has been saturated to an activity of 1 curie/g, whereas with less favorable structures the activity is frequently limited to millicuries or microcuries per gram. The diffusion method has successfully induced measurable, though often small, permanent krypton activity in all substances tested so far. Therefore the title of "universal" indicator is used. Uniform incorporation of krypton into a solid is possible if a fine powder is saturated with krypton and then compressed into a solid mass.

The temperature stability of the kryptonates varies. Some lose most of their activity at atmospheric pressure when at temperatures of 75 to 100°C, whereas copper kryptonate retains 75% of its activity to 900°C. Generally, the stability at room temperature is excellent.

Clathrate Ozone Analyzer. Using the Kr^{85} clathrate, Hommel, Chleck, and Brousaides at Tracerlab developed a very successful analyzer for ozone in the air.[4,15]

Analysis of air for ozone has received increasing attention due to acute problems of air pollution and the associated health hazard. It results in eye and nasal irritations, reduced visibility, injury to plant life, and breakdown of rubber articles. An interest in the determination of atmospheric ozone in weather studies has also developed because it appears that knowledge of ozone-meteorological relations will eventually lead to improved weather forecasting. The principal methods used until now for ozone determination are based on the classical potassium iodide–iodine method, fluorescence, absorption of radiation by ozone, and special methods such as the cracking action of ozone on elongated rubber. Most of the chemical methods are relatively insensitive and nonspecific; for example, the limit of the potassium iodide method is about 1 part in 10^7, and the method is nonspecific. Spectrophotometric techniques require light-path lengths of several hundred feet, and the cracking action of rubber, although highly sensitive, is not specific or continuous. Thus there has long been a need for a portable, lightweight, rugged, and specific ozone analyzer.

Specificity, sensitivity, and simplicity of design are inherent in the clathrate ozone analyzer system, which is based on the reaction given above. The chief limit on the sensitivity is the specific activity of the radioactive salt. In some evaluation tests, when no effort was being made to get maximum sensitivity, 1 part of ozone in 10^{10} parts of air was detected. The specificity is due to the high oxidation potential of the hydroquinone-quinone couple. Only a few oxidizing agents can liberate krypton from the clathrate, and these do not include the airborne contaminants sulfur dioxide and hydrogen sulfide. The clathrate responds to water vapor in the air by dissolution of cage bonds, but the gas can be dried without changing the ozone concentration. Alternatively, the stream can be split, with both ozone and water being measured in the first stream and only water in the second (after destruction of ozone by a silver foil catalyst). Ozone then can be determined by difference.

When the clathrate analyzer is used, 100 to 200 mg of Kr^{85} clathrate is placed in a 2- to 5-mm-diameter capillary (the amount of clathrate and dimensions of the capillary are not critical). The liberated Kr^{85} is passed into a 50-ml chamber containing a Tracerlab TGC-6 Geiger-Mueller (G-M) tube. Counts are observed on a Tracerlab SC-51 Autoscaler. Associated apparatus includes an automatic graphic recorder and rate meter.

One of the most difficult problems in the development of methods for the determination of traces of gases is the preparation of standard samples for analysis. The Tracerlab workers, after trying several methods for preparing air samples containing small concentrations of ozone, decided on the use of ultraviolet ozone lamps, which are sold for sterilization purposes. These lamps, after a break-in period and under carefully controlled conditions, gave an ozone concentration that varied less than 5% during a given day's operation. Use of the proper flow rate and combination of bulbs permitted ozone concentrations between 1 and 500 ppm to be obtained and analyzed chemically by the potassium iodide–iodine method.

The response of the clathrate analyzer to changes in ozone concentration was determined by connecting the ozone-air stream to a source of dry nitrogen, which could be adjusted to give instantaneous changes in concentration. Ozone was passed over the clathrate, and, after

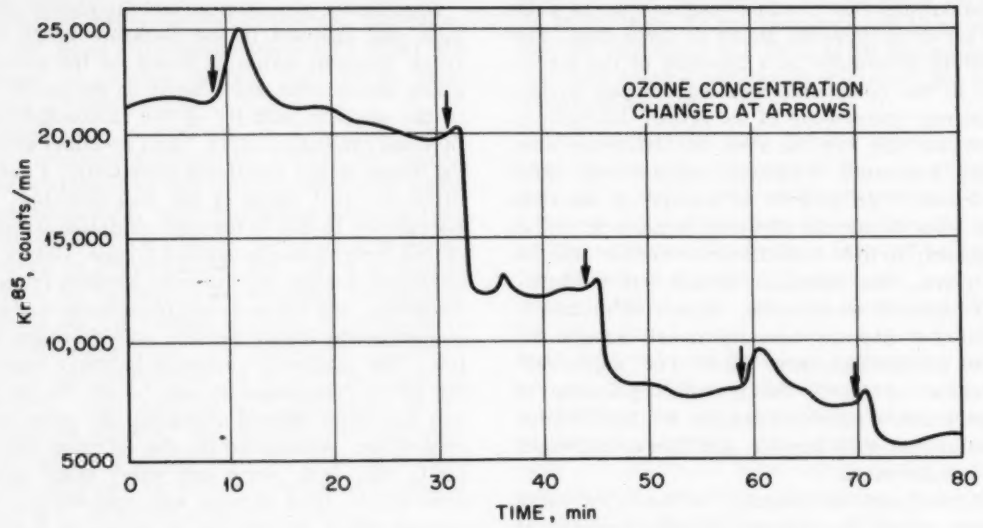


Fig. III-1 Response of ozone analyzer to changes in concentration.[4]

steady-state conditions were reached, the ozone concentration was dropped by increasing the proportion of diluent gas. After a new steady state was reached, the same procedure was again followed for several different, and unmeasured, ozone concentrations. The plateaus on the curve (Fig. III-1) indicate regions of different ozone concentration, and the arrows, the points of dilution.

The 1 min between ozone dilution and counting response is about the same as the time lag due to dead volume. Thus the clathrate reaction very nearly follows the change in ozone concentrations, the response being for all practical purposes immediate. No complete explanation has been given for the rise in the curve immediately after a change in concentration; however, the surge may result from a stripping action of the gas stream, caused by flow-rate adjustments, on loosely sorbed krypton molecules on the surfaces of the clathrate.

To study the variables in the ozone-clathrate reaction, a clathrate cell was installed in a 2-liter bell jar fitted with a manometer (Fig. III-2). A pump of the type used by the U. S. Weather Bureau in sonde balloons circulated

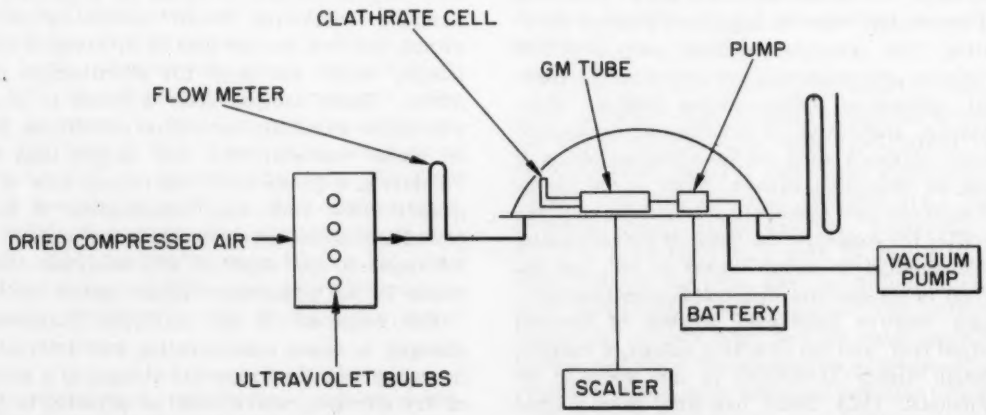


Fig. III-2 Apparatus for environmental ozone studies.[4]

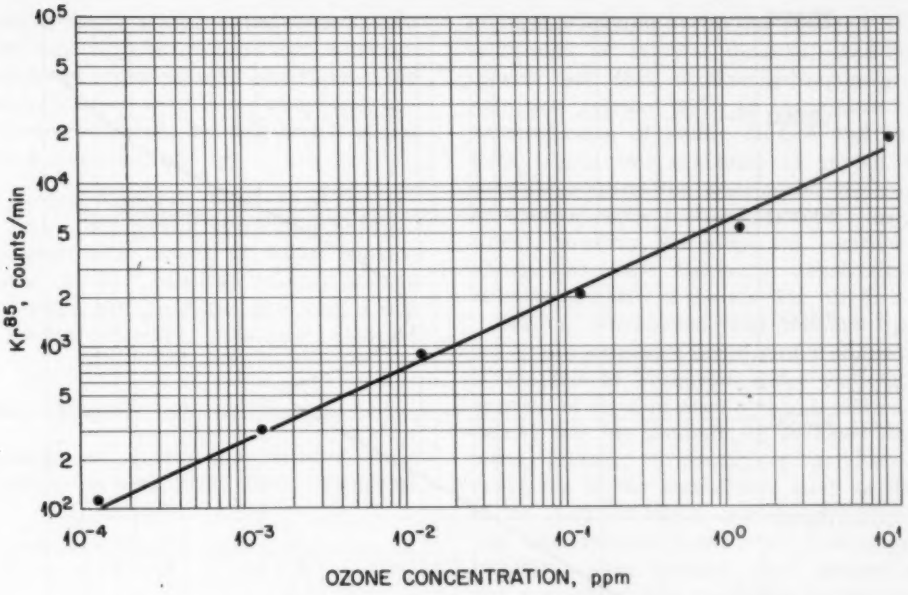


Fig. III-3 Clathrate analyzer response to ozone.[4]

the gas through the clathrate cell and into the detector. For static tests, the chamber is pumped to a high vacuum and ozonized air is admitted. After the system is closed, a pump circulates the gas through the clathrate and detector and returns the gas into the chamber. Although deozonized air and Kr^{85} are returned to the chamber, the interval for data gathering is small and the chamber volume is large enough to maintain an almost constant concentration during the measurement. In dynamic measurements the observation period is unlimited since a fresh stream of air and ozone is constantly fed to the chamber. Flow is maintained with a vacuum pump, and desired pressures are controlled by valves located at feed and exit ports.

The static system was used to test the response of the clathrate cell to a wide range of ozone concentrations at atmospheric conditions. The chamber was filled with 13 ppm ozone at 1 atm, and the counting rate of the Kr^{85} was taken. The chamber was then pumped to 76 mm Hg and repressured with dry nitrogen to 760 mm Hg, where the counting rate was again measured. Thus a given concentration of ozone was successively diminished by increments of 0.9, and an ozone concentration range of five

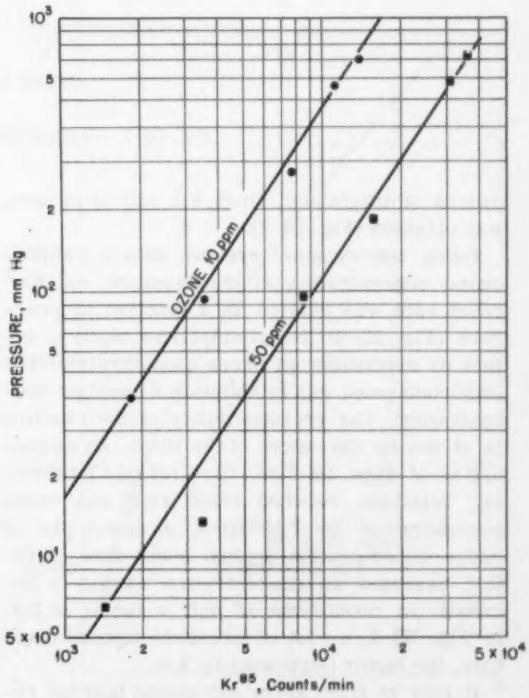


Fig. III-4 Pressure dependence of ozone-clathrate reaction.[4]

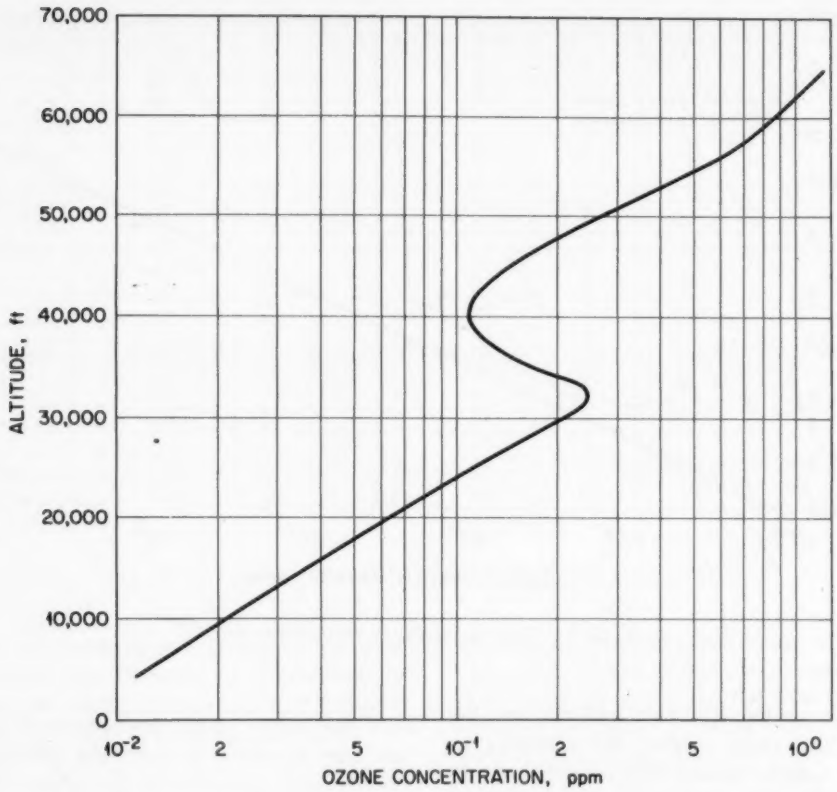


Fig. III-5 Vertical profile of atmospheric ozone.[4]

orders of magnitude, from 0.1 ppb to 10 ppm, was obtained (Fig. III-3).

Using the dynamic system, with a constant ozone concentration in the chamber, the Kr^{85} count rate was studied as a function of pressure (Fig. III-4). No attempt was made to select or determine an ozone concentration. The important point was to maintain a constant concentration. The reproducibility of the reaction is shown by the nature of the slope. An examination of Figs. III-3 and III-4 reveals interesting relations between count rate and ozone concentration. In Fig. III-3, a direct plot of ozone concentration against count rate, a 10-fold decrease in concentration causes a decrease in count rate of only a factor of 2.9. In Fig. III-4, a plot of pressure against count rate, the factor decreases by 4.8.

It may at first seem surprising that the reaction efficiency should increase with a reduction of pressure or ozone concentration. However, the reaction does not involve krypton directly and indicates only the breaking of bonds to release the gas. The rupture of any cage bond will release a krypton molecule, whereas the rupture of two bonds on the same cage will still only release one molecule. Thus, if the ozone density about the clathrate is reduced, either by a pressure or concentration reduction, the probability that two or more ozone molecules will inefficiently effect the release of the same gas molecule is diminished.

Following these and other preliminary tests, the analyzer was tested in balloon flights. Two flights were made using conventional meteorological equipment. In the first flight, data were obtained on the temperature profile and radiation background between sea level and 75,000 ft. This information was needed to aid in interpretation of data from the ozone analyzer since the ozone-clathrate reaction is temperature sensitive and the final measurement of the radioactivity of the evolved Kr^{85} requires a knowledge of the background.

The test of the analyzer in the second flight went quite smoothly up to 75,000 ft. Although the signal-to-background ratio was bad and the ozone concentrations were lower than those obtained by other methods, the data profile shown in Fig. III-5 was obtained.

Sulfur Dioxide Analyzer. Workers at Tracerlab[3,16,17] have also developed an effective analyzer to determine traces of sulfur dioxide in air using the Kr^{85} clathrate. Sulfur dioxide will not oxidize hydroquinone; however, it will oxidize sodium chlorite, releasing chlorine dioxide:

$$SO_2 + 2NaClO_2 \rightarrow 2ClO_2 + Na_2SO_2$$

(This equation is an oversimplification since experimentally it was found that 1 molecule of sulfur dioxide released between 4 and 8 molecules of chlorine dioxide.) The chlorine dioxide will then oxidize hydroquinone clathrate and release Kr^{85}. An instrument for the determination of sulfur dioxide was designed using these two reactions in series. In the original tests of the instrument, the response to varying concentrations of sulfur dioxide was linear; however, the slope and position of the entire line were displaced, depending on the relative humidity of the air (Fig. III-6).

Bersin, Brousaides, and Hommel[17] carried out a very extensive investigation of this method of analysis at Tracerlab under the joint sponsorship of the Los Angeles Air Pollution Foundation and the Division of Isotopes Development, USAEC. A number of variables in the operation of the instrument, such as the size of the reaction tubes, quantity of sodium chlorite and clathrate in the tubes, flow of gas, and humidity, were studied. They showed that it was possible to pick conditions for the analyzer in such a way that, for the lower sulfur dioxide concentrations, the counting rate did not depend on relative humidity (Fig. III-7).

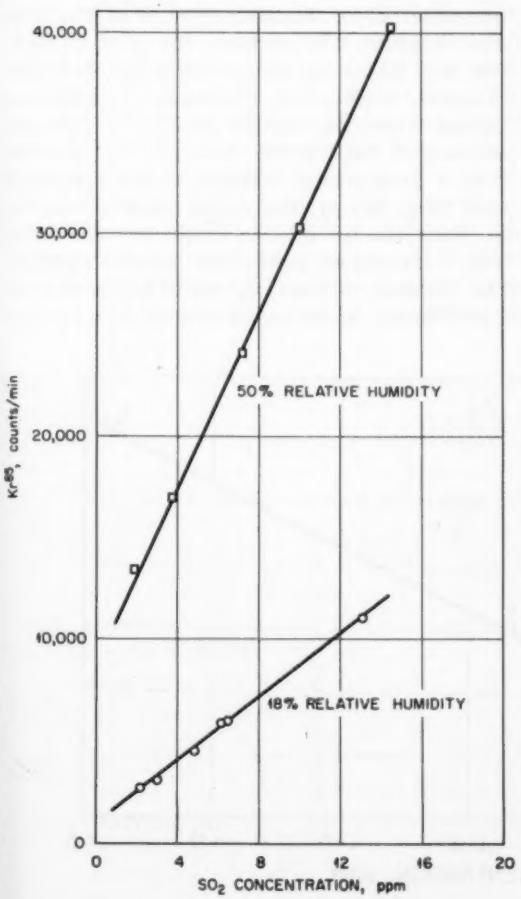


Fig. III-6 Effect of humidity on counting rate.[17]

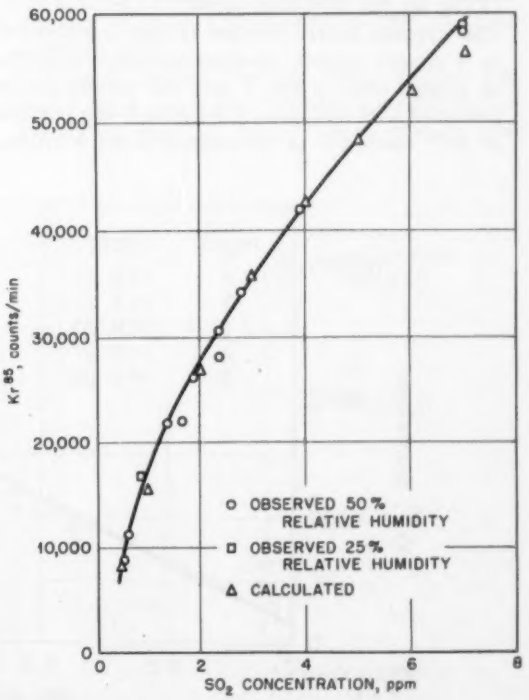


Fig. III-7 Calibration curve for sulfur dioxide at two relative humidities.[17]

In an eight-day test to determine the stability in continuous operation, the data shown in Fig. III-8 were obtained. The only gases that should interfere with the operation of this analyzer are those which can oxidize either sodium chlorite or the hydroquinone in the Kr^{85} clathrate. The oxides of nitrogen and ozone are the only commonly occurring atmospheric gases that should be any trouble.

On the basis of these tests, an instrument for use in field monitoring was designed. A schematic drawing of this instrument is shown in Fig. III-9.

Kr^{85} Clathrates for the Analysis of Solutions. Rotariu, Hoskins, and Hattori at Booz-Allen[6] developed a technique for determining iron by the release of Kr^{85} from the clathrate. It was hoped that by this technique iron could be determined in the parts-per-billion range. The study was discontinued before this goal had been achieved; however, it was shown that it was possible to make determinations in the 2- to 20-ppm range.

In this method, ferric ion is extracted into trifluorotoluene with thenoyltrifluoroacetone (TTA) as the chelating agent at a *p*H of 2 to 3. The organic ferric solution is then transferred to a closed system, gaseous hydrogen chloride is passed over it for 8 sec, and excess gas is removed with nitrogen. A weighed 1-mg portion of Kr^{85} clathrate is then added to the solution, and it is stirred for 15 min. The reactants are frozen in Dry Ice–acetone, and the Kr^{85} is pumped into an ionization chamber and counted.

Although this method will determine 2 to 20 ppm iron in the trifluorotoluene-TTA solution, the results of studies on the distribution coefficient of ferric ion between water and this organic phase indicate that three to four extractions would be needed to approach quantitative extraction. Also, no results of tests on the determination of iron in aqueous solution using the entire procedure are given; therefore it is impossible to judge the overall precision and accuracy possible with such a method.

Determination of Active Hydrogen with Lithium Aluminum Tritide. Krynitsky, Johnson, and Carhart[18] in 1948 published a method for determining active hydrogen in such compounds as water, alcohols, and acids by reaction with lithium aluminum hydride at 0°C in an ether solution. The pressure change in the system was measured to determine the hydrogen released. Workers at Tracerlab[3,19] substituted lithium aluminum tritide for the hydride and determined the tritium released by counting with a proportional counter. In the apparatus used (Fig. III-10), the 25-ml reaction cell, 6.5 in. long and 20 mm in diameter, was fitted with a graduated pipette for sample addition. The reaction mixture, 15 ml of approximately 0.2*M* lithium aluminum tritide in diethyl car-

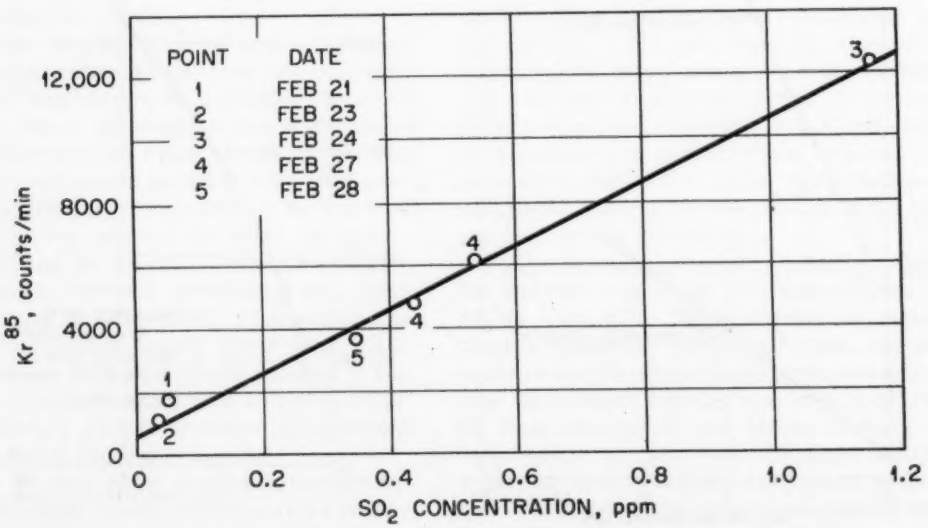


Fig. III-8 Stability of analyzer over an eight-day measuring period.[17]

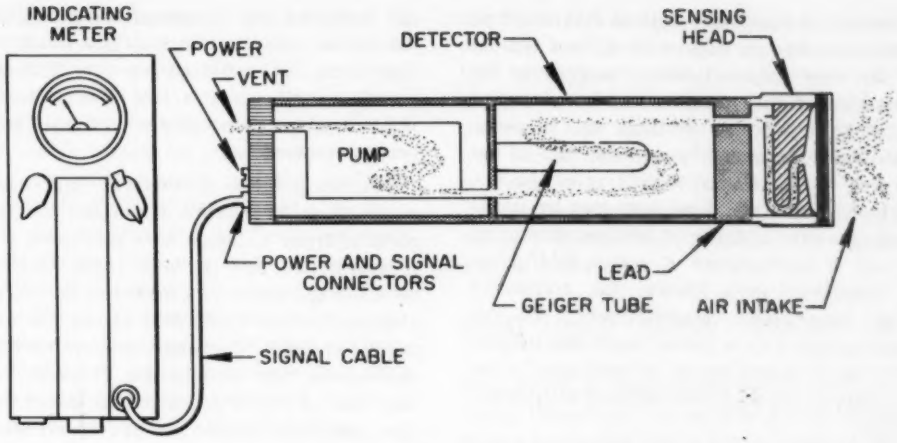


Fig. III-9 Commercial isotope air-pollution monitor.[3]

bitol, was introduced into the reaction cell. The pinch clamp was closed, and counter gas was bubbled through the cell at a flow rate of 40 cm^3/min to obtain the background count. The sample diluted in tetrahydrofuran was introduced, and the tritium was counted for a 10-min period. Fine adjustment of counter gas pressure was made with a screw clamp between the reaction cell and the flowmeter. A bypass line was provided around the reaction cell to allow for the adjustment of flows while reagents were in the reaction cell. The tritium detection equipment consisted of a Tracerlab pulse preamplifier, amplifier, and "1000" scaler; the counter gas was 90% argon and 10% methane.

The value of the method is increased because it is not necessary to prepare a solution of a known specific activity of lithium aluminum tritide in diethyl carbitol. However, the specific activity must be high enough to yield adequate counting rates.

Normal butanol, benzoic acid, aniline, and water were analyzed, different specific activities being used for each. In each case the plot

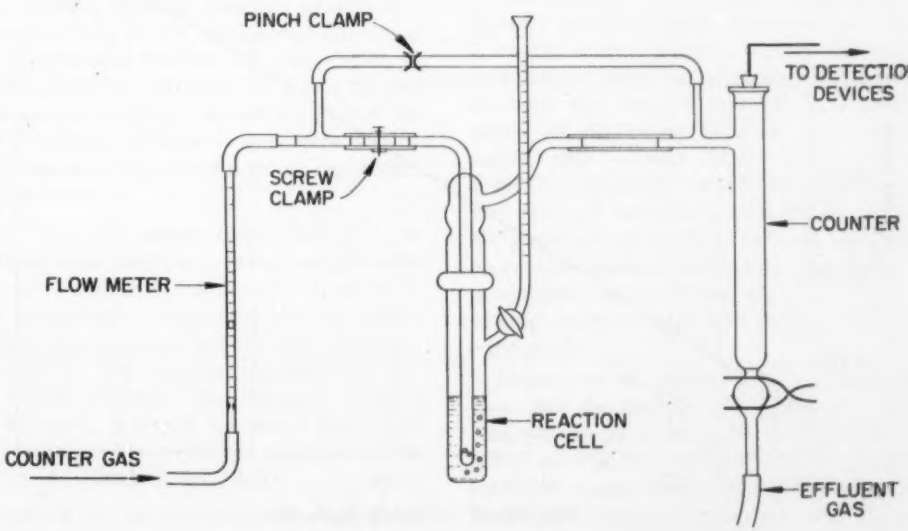


Fig. III-10 Apparatus for active hydrogen determination.[3]

of the amount of compound against counting rate of tritium was linear. Figure III-11 is a typical plot of the data obtained with water. Since the stoichiometry of the reaction had been studied by Krynitsky et al., no attempt was made to correlate data between compounds. Any of the four curves could be used to determine the active hydrogen content of an unknown by using the same specific activity of lithium aluminum tritide; or, if the number of active hydrogens in the compound was known, the compound could be determined quantitatively. All the plots extrapolated to a point below the origin, probably due to fixed losses of hydrogen in the system. This error could be reduced with better cell design.

The sensitivity of the method is very high. The lithium aluminum tritide used in this study had a specific activity of 2.5 mc/mg but was diluted with lithium aluminum hydride by a factor of 10^5. The ultimate theoretical sensitivity with lithium aluminum tritide without dilution would be of the order of 10^{-8} mmole of active hydrogen compound. For practical purposes the sensitivity attainable is limited only by the ingenuity of the analyst in devising more efficient techniques for introducing small sample volumes and a means for sample protection from air moisture. Fifty or more samples may be analyzed on one filling of the reaction cell. In practice the cell should be refilled after one-half the available tritide has been consumed.

Following these successful experiments, the workers at Tracerlab attempted to change the method from a liquid to a gas-solid reaction.[5] It was hoped that the gas-solid reaction could be made the basis of a detector for water vapor similar to that described above for ozone and sulfur dioxide. The experiments were not successful on the microscale because the solid reaction products formed a film over the lithium aluminum tritide and prevented further reaction, but, on a semimicroscale, the reaction proceeded more smoothly and reproducibly. Because of this film formation, however, work with lithium aluminum tritide was discontinued, and exploratory experiments with other reactions were started.

Determination of Dissolved Oxygen in Water. A new analytical technique for determination of dissolved oxygen in water was recently described by Richter and Gillespie of the

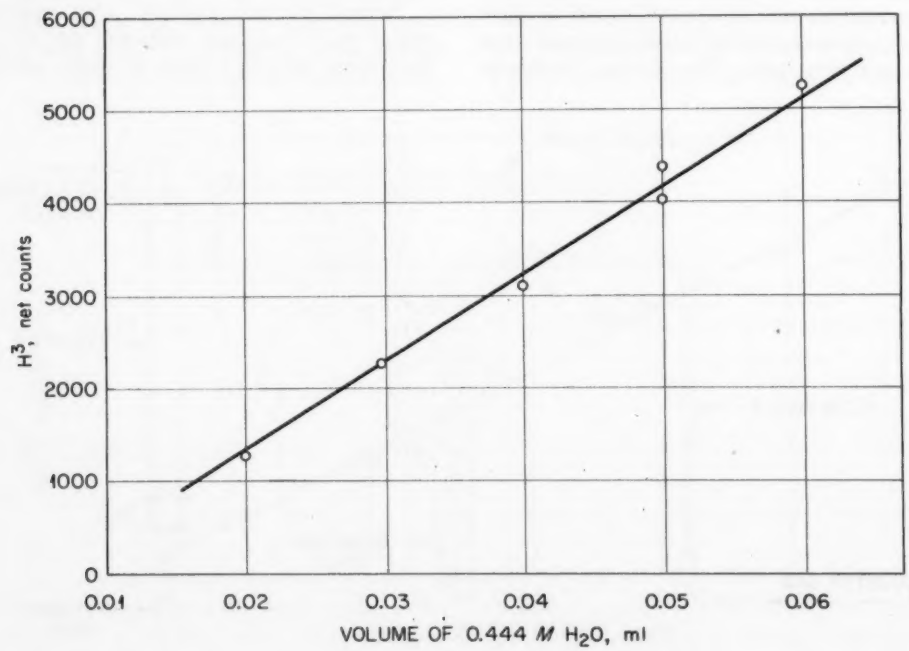


Fig. III-11 Determination of H_2O in tetrahydrofuran.[3]

Research Triangle Institute.[7] The method is based on the rapid and quantitative oxidation of thallium metal by oxygen dissolved in water, even in extremely low concentrations. This fact, discovered by Wright and Lindsay,[20] had been used in an industrial instrument in which the water is passed over thallium and the electrical conductivity of the effluent stream is measured. By substituting Tl^{204} and counting the effluent stream, Richter and Gillespie determined 0.2 ppm oxygen in pure water and used the method for water containing many more impurities than in the original conductivity method. The authors believe that this method is potentially more sensitive than any other known method for this determination.

The analyzer is a 10-mm-diameter 20-cm-long glass column filled with radioactive thallium metal electrodeposited on copper turnings and a flow type G-M tube. The sample is introduced at the top of the column and flows slowly through it. Thallium-204 ions released by the reaction of dissolved oxygen with the thallium enter immediately into the G-M tube and are detected. The Tl^{204} counting rate is a direct measure of the oxygen concentration.

For oxygen concentrations in the parts-per-million range, the radioactive thallium (4480 mc per gram of thallium) was diluted with inactive thallium metal so that the final activity was 2.04 mc per gram of thallium. The Tl^{204} has a long enough half-life (3.6 years) that decay over several months does not greatly reduce the sensitivity of the technique. Also, since Tl^{204} is a pure beta emitter, the glass walls of the column provide sufficient shielding that no radiation hazard exists. It is estimated that the analyzer, excluding scaling or recording equipment, can be built for about $100 in equipment and isotope costs.

Waters of known oxygen concentration were prepared by equilibrating a water sample with nitrogen and oxygen mixtures. Flow rates of the two gases were measured, and the gases were mixed and then bubbled through water for several hours. The sample so prepared was passed slowly through the thallium column (1 to 3 ml/min). A known volume of water was removed from the reservoir for analysis by the well-known Winkler method.

Response of the oxygen analyzer to waters of markedly different salt contents with varying oxygen concentrations is shown in Fig. III-12; the count rate is corrected for dead-time losses and for backgrounds and is plotted as a function of oxygen concentration. The linearity for oxygen in distilled water is excellent, but departure from linearity was noted for sea water and mock sea water containing only sodium chloride.

Thallium determinations on the effluent solutions indicate that oxidation of thallium is stoichiometric:

$$4Tl(s) + O_2(aq) + 2H_2O(l) \rightarrow 4Tl^+(aq) + 4OH^-(aq)$$

One gram of oxygen liberates 25.6 g of thallium; therefore 1 g of thallium is sufficient to assay approximately 5 liters of water saturated with air. The column retains its calibration from beginning to end, and the thallium is completely inert to oxygen-free water.

The sea water used in this study contained 17.7 g of chloride per liter. Thallium analysis of the effluent solution from the column indicated that count rate corresponded to liberated thallium, but this amount did not agree with the expected quantity from the influent oxygen solution except at low oxygen concentrations. Mock sea water consisting of 29.6 g of pure sodium chloride per liter of distilled water gave results similar to those for sea water. Sodium chloride thus appears to cause the departure from linearity.

The results of several experiments performed in an attempt to understand the nonlinearity in sea-water solutions indicated that interaction of thallium, copper, and oxygen was causing the loss of thallium. Columns prepared by depositing thallium on a metal less active than copper should give satisfactory linearity. Although linear response by an instrument is desirable, nonlinear action does not preclude its usefulness in any applications. Since the thallium release in sea water is reproducible, if not linear, this technique can be a good analytical tool once the column has been calibrated.

Studies on the effect of the solution *p*H indicate that the method is suitable for solutions with *p*H values between 5 and 10 and that acidic solutions can be analyzed if the *p*H is brought into this range with a suitable buffer. Nitrate salts interfered, but chromates did not at *p*H 8.

The sensitivity of the column with a specific activity of 2.04 mc per gram of thallium is

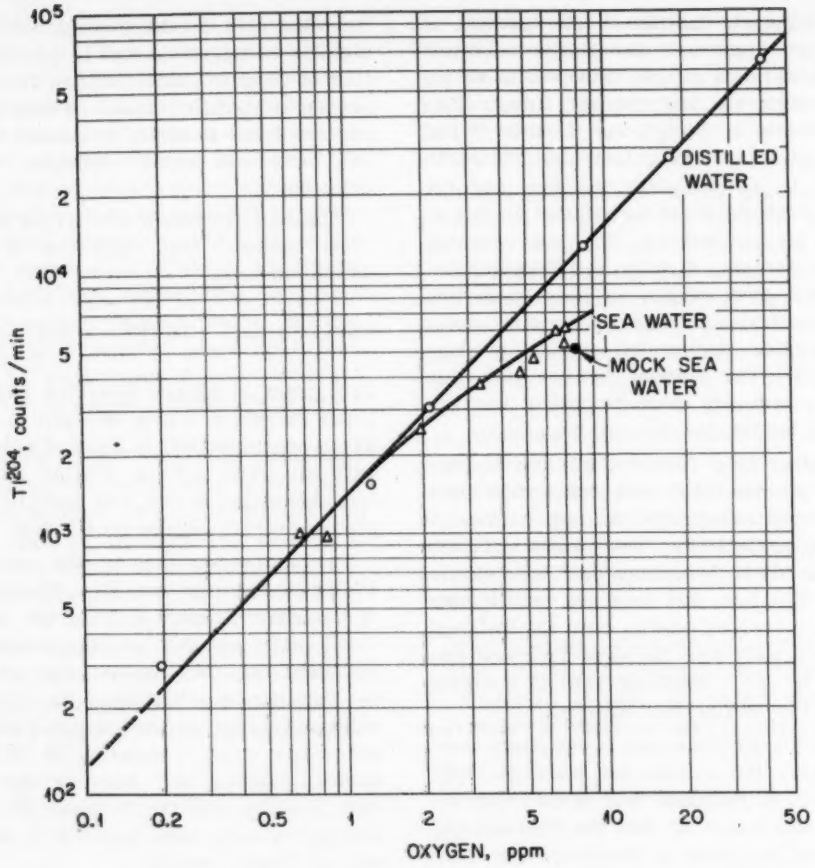
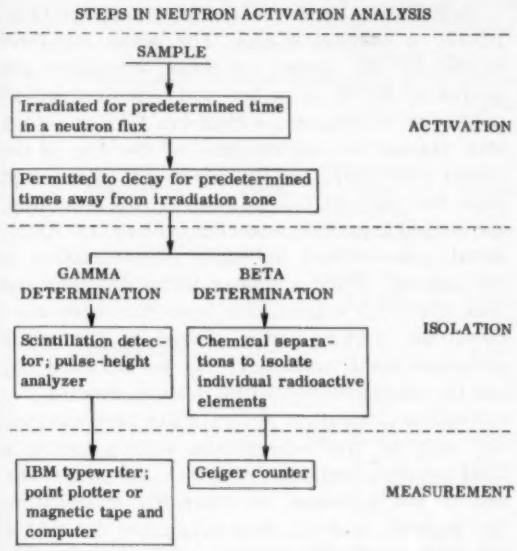


Fig. III-12 Response of Tl^{204} analyzer to dissolved oxygen in distilled water and sea water.[7]

about 0.2 ppm, which produces a Tl^{204} counting rate equal to the background counting rate of the detector. If the 4478 mc/g material obtained from Oak Ridge National Laboratory (ORNL) for the study had been used without dilution, the sensitivity of the column would have been 0.1 ppb. (The ORNL Isotopes Catalog lists Tl^{204} with a minimum specific activity of 1000 mc per gram of thallium.) Oxygen concentrations in this range were not investigated because of time limitations and the difficulty of obtaining solutions of known parts-per-billion oxygen concentrations. Sensitivity could be increased by at least a factor of 10 by using a larger counting tube. Still another factor of 10 in sensitivity should be attainable by a reduction in the background counting rate, which was about 100 counts/min. Thus a sensitivity of about 1 part of oxygen per trillion parts of water should be practical. This speculation is based on the assumptions that metallic thallium is totally insoluble in oxygen-free water and that radiolytic decomposition of water would produce no more oxygen than this.

Neutron Activation Analysis

The basic principles of activation analysis are very simple. A sample of material is bombarded by some type of particle, usually neutrons, which produces radioactivity in either the matrix material, the impurities, or both; the activity may or may not be isolated; and the type and amount of the activity are determined. An outline of the general steps in neutron activation analysis was given recently by Aebersold:[21]


STEPS IN NEUTRON ACTIVATION ANALYSIS
SAMPLE
Irradiated for predetermined time in a neutron flux
ACTIVATION
Permitted to decay for predetermined times away from irradiation zone
GAMMA DETERMINATION
BETA DETERMINATION
Scintillation detector; pulse-height analyzer
Chemical separations to isolate individual radioactive elements
ISOLATION
IBM typewriter; point plotter or magnetic tape and computer
Geiger counter
MEASUREMENT

Neutron activation analysis was originated in 1936 by Hevesy and Levi,[22] who exposed samples of rare earths to a radium-beryllium neutron source to detect and determine impurities. The method, however, did not gain wide acceptance until after 1946 when nuclear reactors became available to provide high-flux sources of neutrons.[23] Since then many analytical methods using neutron activation have been developed, but the majority of these are suitable only for laboratory use since they require extensive radiochemical separation and take too much time for consideration in process control. Neutron activation is now moving out of the research laboratory and into broad applications throughout science and industry, mainly because of the development of new techniques and methods.

Meinke and Shideler[24] have suggested that activation analysis will come into general use only when neutron sources suitable for use in an individual laboratory are available and when it is possible to complete an analysis in a matter of minutes (or at the most 1 hr) instead of days. Current research in this field is directed toward solution of both these problems.

Although there have been many papers published on activation analysis, no textbook or monograph has appeared; however, several such books are said to be in preparation. "Source Material for Radiochemistry"[25] gives a reasonably good bibliography of the most important papers in the field up to 1960. Gibbons et al. have published two bibliographies,[26,27] and Koch has compiled a handbook.[28]

The USAEC is continuing its strong interest in activation analysis by supporting a broad range of research and development projects, including preparation and evaluation of isotopic neutron sources; testing of small accelerators for production of neutrons; activation with fast neutrons; development of rapid radiochemical techniques for isolation of radioisotopes; use of multichannel gamma-ray spectrometers for determination of activities; application of instrumentation and computer techniques to provide rapid, automatic, and routine analyses of very large numbers of samples; and use of these techniques in such diverse applications as analysis of the ocean bottom, the surface of the moon, and samples of interest in criminal investigations. Some of these newer developments are reviewed here.

Neutron Sources. Olive, Cameron, and Clayton of the United Kingdom Atomic Energy Authority (UKAEA) Isotope Research Group recently reviewed high-intensity neutron sources and their applications in industry.[29] Nuclear reactors are not considered, but radioisotope sources and accelerating machines are reviewed in detail.

1. Isotopic Neutron Sources. The UKAEA report concludes that radioisotope sources such as those shown in Table III-1 will usually be limited to a neutron yield of 10^6 to 10^7 neutrons/sec. If higher source strengths are needed, small accelerators or reactors will probably be more economical.

Hennelly[30] published information on a 1000-curie Sb^{124}-beryllium source that has a neutron yield of 10^{10} neutrons/sec. This source has a half-life of only 60 days, but it can be regenerated easily in a reactor. Hennelly suggests that two sources be prepared, with one

Table III-1 RADIOISOTOPE NEUTRON SOURCES

Source	Half-life	Yield, neutrons/(sec)(curie)
Pb^{210}-Be(α,n)	22 years	2.3×10^6
Ac^{227}-Be(α,n)	22 years	1.5×10^7
Ra^{226}-Be(α,n)	1622 years	1.3×10^7
Po^{210}-Be(α,n)	138 days	2.5×10^6
Th^{228}-Be(α,n)	1.9 years	2.0×10^7
Sb^{124}-Be(γ,n)	60 days	1.0×10^6

in the reactor being reactivated while the other is in use. (The Sb^{124}-beryllium source is the only one shown in Table III-1 that can be reactivated.) He estimates that sources could be operated for $5000 per year by this method. The British review[29] points out that the worst drawback from this type of source is its intense associated gamma-ray emission, about 10 r/hr at 1 ft from 1 curie.

Strain and Leddicotte[31] recently published a description of alpha, gamma, and neutron sources made from Am^{241}. They prepared Am^{241}-beryllium sources that had a total neutron emission of 4.5×10^6 neutrons/sec, resulting in a flux of 2.36×10^4 neutrons/(cm^2)(sec). Although this flux is low, they believe it would be suitable for determination of macro concentrations of a large number of materials. The Am^{241} was >99% pure radiochemically, containing ~0.1 wt.% plutonium. However, the 16 wt.% stable contaminants (9% rare earths, 3% iron, 2% chromium, and 2% sodium-nickel-copper) had to be removed to provide maximum specific activity of the sources. This was done by a modification of a Dowex-1 resin ion-exchange process,[32] with elution by 5*M* NH_4SCN. The americium was extracted from the eluate into di-2-ethylhexyl phosphoric acid (D2EHPA), with decontamination factors of at least 10^5 from the rare earths, plutonium, and sodium, and the Am^{241} recovery was ~90%. The 10% loss occurred in the ion-exchange-column separation because Am^{241} eluted only very slowly with any eluting agent. When the same column was used for processing additional amounts of americium, the retained americium was slowly eluted, at the rate of about 100 μg per liter of 5*M* NH_4SCN.

Two types of neutron sources were produced: americium-boron and americium-beryllium. The Am^{241}-beryllium sources were prepared by thoroughly mixing $Am(NO_3)_3$ in weak HNO_3 (*p*H 2) with 325-mesh beryllium powder and evaporating the mixture to dryness. The dry product was fired at 500°C to expel NH_4NO_3 and convert the americium to AmO_2. The Am^{241}-boron sources were prepared similarly, except that the fired AmO_2-boron powder was combined with 0.050 g of paraffin dissolved in CCl_4 and allowed to dry. The mixture was ground and pressed to a 0.72- by 0.73-in. right cylinder under a static load of 10 tons, and the cylinder was heated to 500°C to remove the paraffin binder.

Both pellets were sealed in containers (Fig. III-13) in several stages. The pellet was first placed in the nickel, or inner, container and heated to 275°C on a hot plate. With standard soldering techniques, a uniform film of solder was placed on the threads of the cap of the nickel container, and the cap was then screwed onto the body of the container. After cooling, the sealed container was cleaned with 1*N* HNO_3, dried, and checked for alpha contamination on its surface. When a smear technique removed less that 75 α counts/min from the outer surface, the source was placed in the outer stainless-steel container, its lid screwed on, and the container sealed by Heliarc welding.

Neutron activation analysis has been carried out with an Am^{241}-beryllium source having a total neutron emission of 4.54×10^6 neutrons/sec. When enclosed in a paraffin moderator, the highest neutron flux attainable is 2.36×10^4 neutrons/(cm^2)(sec) as measured by gold, manganese, and cobalt foil activation. The sensitivities of a number of elements have been tabulated with a series of pure oxides, nitrates, and carbonates, for various irradiation times, using a 3- by 3-in. NaI detector coupled to a 20-channel pulse-height analyzer for discriminatory counting. Table III-2 gives a partial listing of these elements. Although these neutron sources are not sufficiently intense to allow trace analysis for more than a few isolated elements, they offer a rapid and specific analysis method for macroelement concentrations in a wide variety of materials.

2. Accelerator Neutron Source. The use of accelerators for production of neutrons was recently reviewed in *Nucleonics*.[33] Currently the chief interest in neutron sources for activation analysis seems to be in the small particle accelerators that generate neutrons

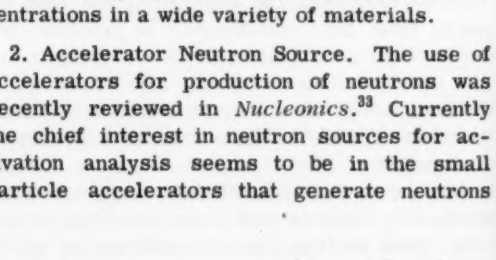


Fig. III-13 Am^{241}-beryllium neutron-source container.

Table III-2 SENSITIVITIES OF A FEW ELEMENTS FOR NEUTRON ACTIVATION WITH AN Am^{241}-BERYLLIUM NEUTRON SOURCE

[Thermal Flux: 2.4×10^4 neutrons/(cm²)(sec)]

Element	Isotope produced	$t_{1/2}$	Gamma energy, Mev	Irradiation time	Energy, Mev	Activity, counts/(min)(mg)
Hafnium	189m	19 sec	0.161	1 min	0.02 to 0.21	2
Scandium	46	19.5 sec	0.14	1 min	0.02 to 0.21	3
Tungsten	183	5.5 sec	0.105	1 min	0.02 to 0.21	0.01
Aluminum	28	2.3 min	1.78	5 min	1.6 to 1.88	0.04
Iodine	128	25 min	0.455	30 min	0.72 to 1.48	0.08
Silver	108	2.3 min	0.44, 0.60	5 min	0.36 to 0.74	0.2
Magnesium	27	9.4 min	0.84, 1.02	10 min	0.72 to 1.48	0.002
Manganese	57	2.58 hr	0.845	60 min	0.80 to 1.18	0.08
Gold	198	2.70 days	0.412	2.6 days	0.34 to 0.53	3
U^{238}	U^{239}	23.4 min	0.074	40 min	0.04 to 0.42	1
U^{235}	FP's	Short	Many	40 min	0.04 to 0.42	1

by the $H^3(d,n)He^4$ reaction. In the British review mentioned above,[29] data from the *Nucleonics* article on such machines, which cost about $20,000, are summarized.

Two recent reports[34,35] describe the installation and use of Texas Nuclear Corporation Cockcroft-Walton neutron generators. The installation at the University of Michigan[24,34] is routinely producing a thermal neutron flux of 2×10^8 neutrons/(cm²)(sec), whereas ORNL (Ref. 35) generates a fast neutron flux of about 3.5×10^8 neutrons/(cm²)(sec). The generator, which both these laboratories use,[36] consists of three units: the accelerator with vacuum system, a high-voltage power supply, and a control console. These units, interconnected by cables, can produce neutrons immediately upon installation with no further operations necessary except for mounting a tritium target in the target holder. This generator, which costs about $20,000, accelerates deuterons to 150 kv (Fig. III-14), the deuterons strike a tritium target, and the $H^3(d,n)He^4$ reaction takes place, producing 14-Mev neutrons. Both installations are in suitably shielded rooms equipped with pneumatic tubes to introduce samples into the accelerator beam and to move the activated samples into a counter. The ORNL generator is equipped to do only fast-neutron activations. The Michigan generator has a 30- by 30- by 30-in. water tank so that the neutron beam can be moderated with water; it can therefore be operated as either a fast- or thermal-neutron generator.

One of the most difficult problems with this type of generator is the tritium target. The usual commercial target is about 1 curie per square inch of H^3 adsorbed in a layer of titanium about 200 A thick on a 2-mil-thick stainless-steel backing. With a 150-kv stream of deuterons striking this target in a rather small area, it must be cooled, and, even with cooling, damage and depletion of the target are rather rapid. To improve target life, Meinke and Shideler[34] developed a rotating target.

The target assembly (Fig. III-15) gradually rotates a long (10.5 by 2 in.) titanium-H^3 target in front of the beam and gives an even flux of $>10^9$ neutrons/(cm²)(sec) for approximately 22 hr. The strip of target material is clamped to the inner surface of a stainless-steel drum, the active side of the target forming the inner surface of a cylinder 3.5 in. in diameter. The entire back surface of the target is cooled by mercury, which is circulated through a water heat exchanger by a small magnetic pump.

An O-ring seal on a rotary shaft transmits motion from a 24-hr clock drive situated outside the housing to rotate the drum at a rate of 0.00725 in./min; this corresponds to ~200 min for about 95% target use in a beam 1.5 in. wide at 1 ma. The drum target is situated with its axis inclined 45° to the beam axis so that the beam strikes the target through the open end of the drum. This angle also increases the area of the target exposed to the beam, thereby reducing the beam-current densities on the target.

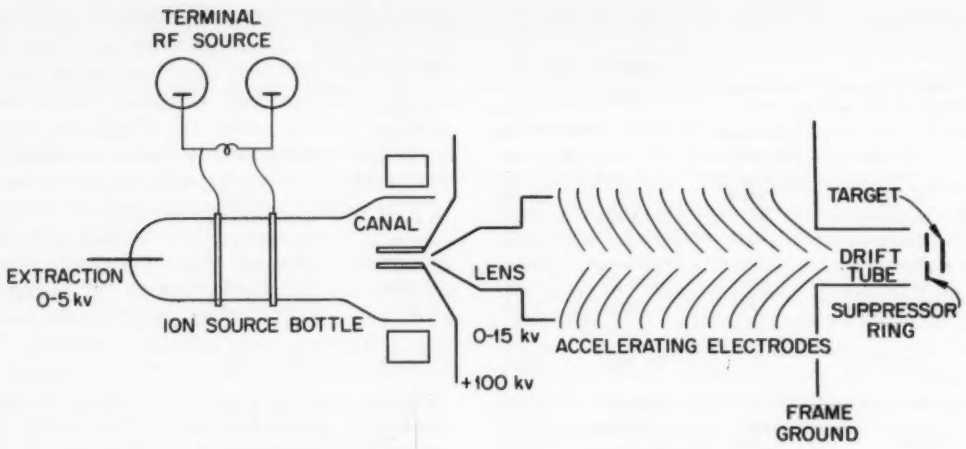


Fig. III-14 Schematic diagram of accelerator beam tube and ion source.[34]

3. Neutron Source for Activation of Flowing Stream. An interesting type of neutron source has recently been described by Gluck, McFarling, Kircher, Townley, and Sunderman at Battelle Memorial Institute[37] in a study done for the USAEC Division of Isotopes Development. Designed for activation analysis of a flowing stream, the irradiator uses a polonium-beryllium source that can be lowered into an activation cell.

A combination of neutron-source storage container and activation-cell holder was built and is shown in Fig. III-16 with the source in the stored position and surrounded with 16.5 in. of borated paraffin deposited in 0.25-in.-thick layers. The paraffin provides adequate shield-

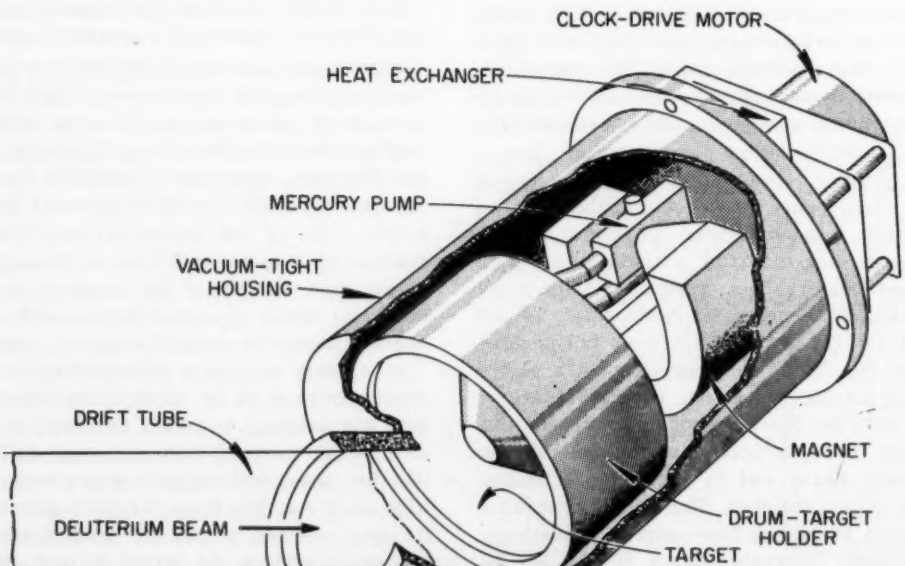


Fig. III-15 Long-lived target assembly.[34]

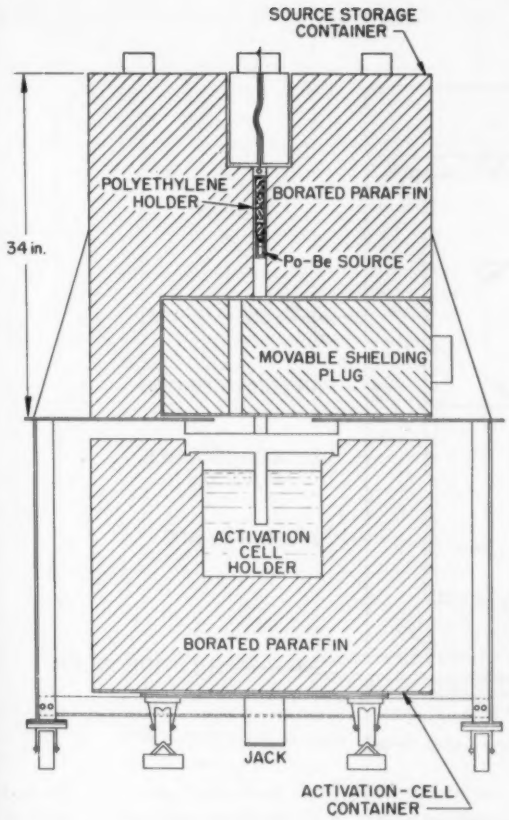


Fig. III-16 Neutron-source storage container and activation-cell holder.[37]

ing for a 50-curie source. Because of their weight the two containers were designed to move individually. An activation cell (Fig. III-17) with a volume of 2400 cm^3 was built to fit in this holder. This cell is constructed of 6-in.-wall stainless-steel tubing, has a thimble suspended in the center, and overflows from the top. In order to achieve good mixing, a diffuser plate above the inlet and two baffle plates were incorporated. The outer surface of the activation vessel was coated with about 1.5 in. of paraffin, and the vessel was permanently installed in the container. Provisions were made for washing and draining the cell. Void spaces around the installed cell were filled with polyethylene pellets for additional shielding.

To evaluate the system, the activation cell was installed in the cell holder. Figure III-18, a schematic of the flow system, shows the activation cell and the gamma-counting cell. The beta-counting cell can readily be substituted. During operation, inactive solution from the stirred process-solution tank was pumped through the system at a constant flow rate, entering through the bottom, overflowing from the top, and then passing through the counting cell and into the storage container. Solutions of four different elements were pumped through the cell, and their induced activity was determined as a function of the flow rate. The flow rates ranged from 50 to 490 ml/min, equivalent to activation times of from 48 to about 4 min.

Activation sensitivities for several elements—manganese, indium, vanadium, and silver—were examined under flow conditions with the 50-curie source. Typical results for vanadium solutions are shown in Fig. III-19. The data plotted are the activities corrected for the different activation times by dividing the observed activity by $1 - e^{-\lambda t}$, where t is the irradiation time. The effective irradiation time was calculated for each flow rate by dividing the volume of the activation cell (2400 ml) by the flow rate. Vanadium-52 with a half-life of 3.76 min was produced.

The corrected activity at the detector was found to decrease with decreasing flow rate, which is indicative of a time delay between activation and counting. Such a delay is inherent in this system for low flow rates or short-lived activities, or both, because of the relatively large counting-cell volumes. The calculated volume necessary to account for the observed results, called "apparent holdup volume," was found to be 675 and 620 ml for data obtained with the gamma- and the beta-counting cells, respectively. It is difficult to account for these exact volumes, but they depend on the counting-cell construction and method of injecting the solution to be counted, the activation-cell construction, and the amount of mixing in these cells. The assumption of this holdup volume, with the resulting radioactive decay as the solution traverses the volume, accounts very well for the results as shown in Fig. III-19 except at low flow rates. It is apparent that mixing of the solution is not good at the low flow rates.

Rapid Methods of Analysis. In addition to small portable neutron generators, routine activation analyses will require more rapid methods. Three different approaches are being investigated: (1) quick radiochemical separa-

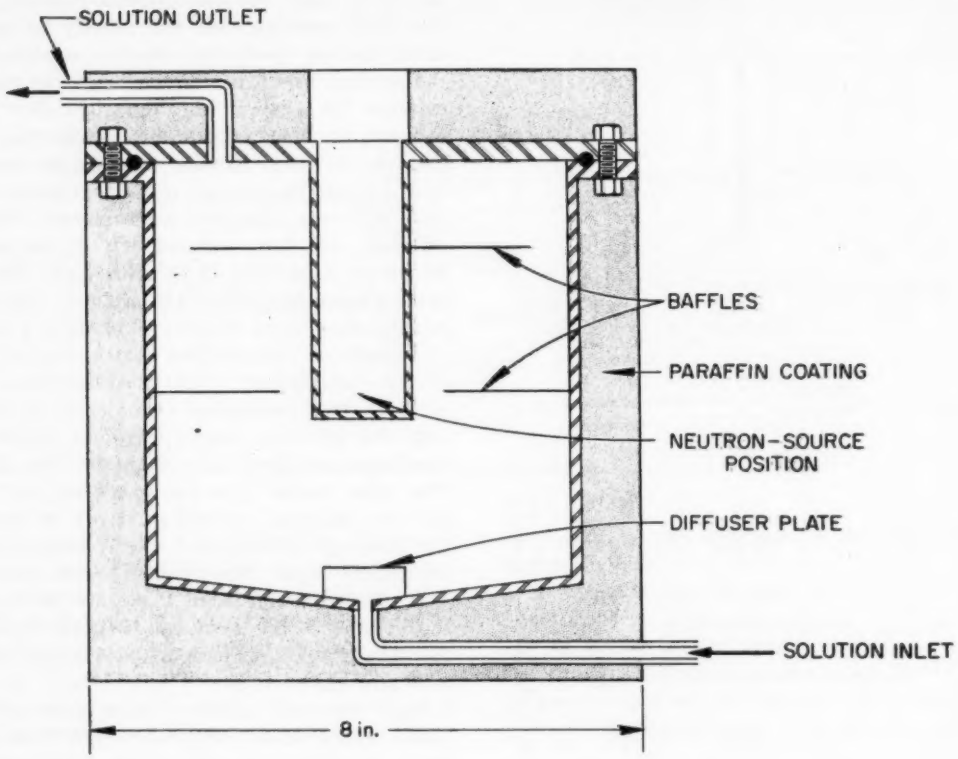


Fig. III-17 Detail of activation cell.

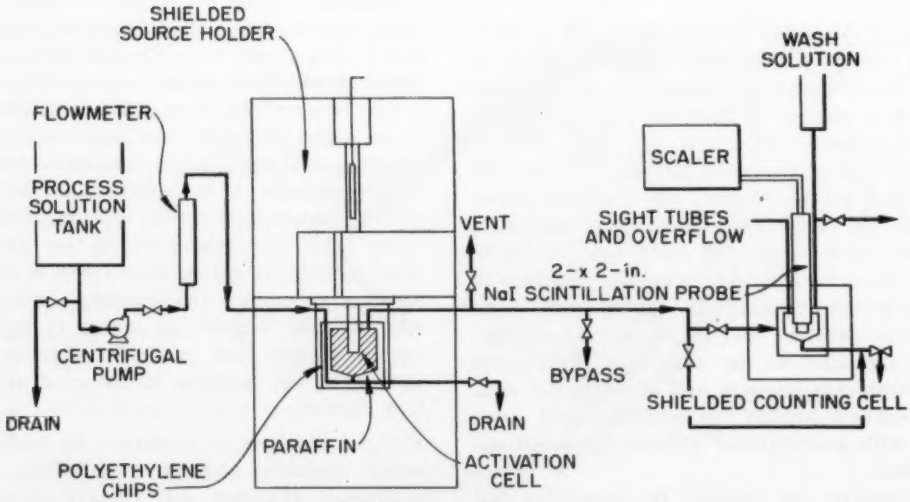


Fig. III-18 Diagram of continuous neutron activation flow system.[37]

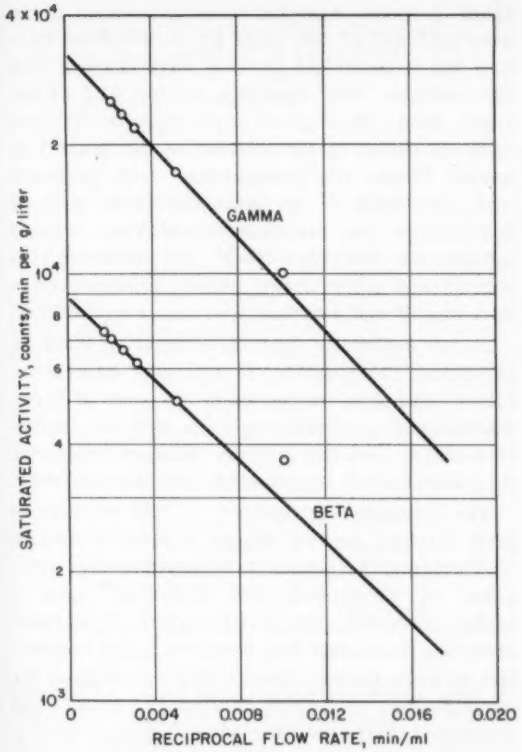


Fig. III-19 γ and β activities of V^{52} produced by activation of vanadium solutions in a flowing system. $V^{52}t_{1/2}$, 3.76 min. Apparent holdup volume, 675 ml for γ and 620 ml for β (Ref. 37).

tion methods following activation so that isotopes with short half-lives can be determined; (2) instrumental methods in which neutron activation is followed by gamma-ray spectrometry with no chemistry, the gamma-ray spectrum frequently being determined several times to measure half-lives of various activities; and (3) very elaborate techniques in which the activation, counting, and calculating sequences are automatically controlled.

1. Fast Radiochemistry. Meinke and his coworkers[38,39] have been working for several years to develop and collect methods for the rapid separation of radioisotopes. Although it is not possible in a brief review of this nature to condense or abstract his 125-page collection of methods,[39] a brief review of the guidelines and philosophy that have gone into the preparation of this collection is instructive and informative:

Radiochemical separations offer one major advantage over standard analytical techniques in that they do not require 100.0% yield. Since in radiochemistry we can determine the yield either by using carrier or by using tracers, we can utilize a much wider spectrum of reagents and procedures than can the classical quantitative analyst. The only limitation is that which is similarly imposed on the synthetic organic chemist, i.e., a separation step must give sufficient yield so that one can go on to the next step.

Unfortunately, those who make use of radiochemical separations, whether in fission-product analysis, nuclear decay schemes, or activation analysis, have too often tended to rely on adaptations of quantitative procedures which are lengthy and involved, to purify the desired radioisotope for measurement. Thus many of the early fission-product procedures were developed in a hurry by combining many individual steps from quantitative work while often only a few steps of a multistep procedure were really effective. These early procedures were of necessity designed rapidly with the information on hand to meet the emergencies of a war situation. Unfortunately many radiochemists today are blindly following these early procedures with little thought as to conservation of time and energy.

Workers in the field of activation analysis seem to be as guilty in this respect as those in other areas. A survey of published articles in this field shows two major classifications. In one group many investigators have sought to eliminate chemical procedures entirely with elaborate techniques of physical discrimination. In the other group the discrimination is chemical but is often quite involved and based usually on rather classical methods of separation.

Dr. Yusuru Kusaka and I have recently completed a rather thorough summary of radiochemical separation procedures reported in the literature for isotopes with half-lives shorter than 20 minutes. Although use of a number of ingenious devices was reported, particularly for isotopes of some of the lighter elements, the preponderance of the procedures was merely "speed-ups" of the standard analytical type techniques such as precipitation, solvent extraction, and ion exchange.

2. Instrumental Methods. In the "instrumental method" of Guinn and Wagner,[40,41] the gamma-ray spectra of samples are determined and compared with the spectra of known samples of simple compounds or elements to show the content of the unknown. The comparison of spectra by hand is a very tedious job, and programs are currently being prepared to do this work on computers. Blotcky, Watson, and Ogborn[42] at a recent symposium gave a paper describing a system using a 400-channel spec-

trometer and an IBM-1620 computer. At the same symposium, Guinn and Lasch[42] described studies being done preliminary to preparing a program for the IBM-7090.

3. Automated Methods. The most elaborate and sophisticated approach to instrumental activation analysis is the work being done at the Activation Analysis Research Laboratory, A & M College of Texas, by Wainerdi and coworkers.[43,44] This work is divided into two parts. In one part an automated system is being designed to activate samples, count the samples with multichannel analyzers, and prepare the data in a form suitable for use on a computer. The second part of the work is the development of suitable computer techniques to resolve the gamma spectra and provide quantitative analytical data.

The original system for automated analysis[45] was designed to operate either by reactor activation of samples in groups of 100 followed by individual and automatic counting or by individual and automatic introduction of the samples into the reactor for activation and automatic counting (Table III-3). With either of these operations, the samples are counted in a 256-channel vacuum-tube gamma-ray spectrometer, and the data are recorded on IBM cards for transfer to the computer. All sample transfers are pneumatic. In addition to its automatic features, this system also has built-in features that allow the operator to change almost any phase of the operation on each sample as it comes through.

The speed of operation in the Mark I-Ia mode is determined by the irradiation and counting times. In a demonstration of operation in the Mark I mode, samples were processed at an average rate of 400 in 24 hr. A downtime record for a period of several days showed that the machine was operating during 97% of the time. Table III-4 gives some results obtained with operation of the system in the Mark I-Ia mode. These are preliminary data, gathered and processed in an automatic mode with no corrections or smoothing processes applied outside the computer itself. The elements aluminum and silver were chosen to illustrate a good result and a rather poor one, respectively.

A new automatic system, designated Mark II, is under construction. It will take bulk irradiated samples, count them on one of three 400-channel transistorized gamma-ray spectrometers, and record the data on magnetic tape suitable for introduction into the computer.

The computer programs in this work have gone through several stages in their evolution. The original activation analysis computer program of Kuykendall and Wainerdi[46] used a highly complex, sophisticated spectrum-stripping technique that required many sequential measurements. This program, designed for an IBM-704, used both gamma-ray energy and measured half-lives for identification. Such a program was not readily adaptable for use in a fully automatic system. The simplified program of Fite, Gibbons, and Wainerdi[47] was therefore developed with the assumption that large numbers of similar samples would be run, and a general identification could be made manually on a few samples. This program has recently been revised again.[45,48]

Uses of Activation Analysis. Many applications of neutron activation analysis were re-

Table III-3 AUTOMATED ACTIVATION ANALYSIS

Mark I-Ia Mode of Operation

1. 100 samples are placed in a plastic storage block
2. The first sample is transferred from the storage block to the reactor
3. After proper irradiation time, the sample is transferred from the reactor to the 256-channel gamma spectrometer
4. The sample is counted for a predetermined time
5. The sample is transferred to a lead storage block, and data are recorded on an IBM card
6. The cycle is repeated for each of the 100 samples

Mark I Mode of Operation

1. 100 samples previously activated in a reactor are placed in a lead storage block
2. The first sample is transferred from the storage block to the 256-channel gamma spectrometer
3. The sample is counted for a predetermined time
4. The sample is transferred back to the lead storage block, and data are recorded on an IBM card
5. The cycle is repeated for each of the 100 samples

Table III-4 ANALYSIS OF STANDARDS BY MARK I-Ia MODE OF OPERATION

Amount of element, mg		
Added	Found	Error, %
	Aluminum (Al_2O_3)	
0.725	0.742	2.3
0.704	0.700	−0.6
0.694	0.685	−1.3
0.699		
0.736	0.709	−3.7
0.715	0.751	5.0
0.656	0.664	1.2
		2.3 (av.)
	Silver (Ag_2O)	
0.158	0.115	−27.0
0.140	0.089	−36.0
0.093	0.110	18.0
0.130	0.067	−47.0
0.149	0.093	−38.0
0.102	0.069	−32.0
0.093	0.097	4.3
0.102	0.109	6.9
		26.1 (av.)

ported[49] at the International Conference on Modern Trends in Activation Analysis held at A & M College of Texas in December 1961. A number of applications were also recently reviewed by Wainerdi and DuBeau in a recent issue of *Science*.[50] Only a few uses for crime detection and for analysis of oceanographic cores will be reviewed at this time.

1. Law Enforcement. Several laboratories have recently started an evaluation of the possibilities of using activation analysis to aid in the examination of evidence for law-enforcement purposes.

Guinn[51] of General Atomics has developed a technique to determine whether or not a suspect has recently fired a gun. The technique involves careful cleaning of the suspect's trigger hand; activating the removed material in a reactor; chemically separating the Sb^{122}, Ba^{139}, and Cu^{64}; and determining these activities by gamma-ray spectrometry. In some cases it is possible to tell not only that the suspect has fired a gun, but also the type of ammunition. Guinn is now cooperating with the Los Angeles Police Department on the application of this method.

Bate, Pro, and Leddicotte[52] of ORNL and the Alcohol and Tobacco Tax Division Laboratory, Internal Revenue Service, after examination of a number of samples of black automobile paint and some soil and soot samples, have concluded that activation analysis would be a useful technique for the forensic scientist to use in identification of these materials. After each specimen of paint had been irradiated for 1 hr in a reactor at a neutron flux of 8×10^{11} neutrons/(cm^2)(sec) and the radioactivity measurements made of the short-lived radionuclides, the paint was allowed to decay for 1 day. Each sample was then re-irradiated for an additional 140 hr to build up sufficient radioactivity of longer lived radionuclides. After this later irradiation, each specimen was allowed to decay for about 6 hr before radioactivity measurements were made. This time was long enough to allow the short half-lived radionuclides in each sample to decay sufficiently so that radionuclides of longer half-lives could be more easily detected and determined. After these measurements the samples were allowed to decay, with occasional radioactivity measurements, until the spectra of the induced radionuclide began to show strongly the longer lived radionuclides. This decay time was 30 hr.

The specimen was again analyzed by gamma spectrometry, and the spectral data were recorded. Additional data were recorded seven days after reactor discharge. Table III-5 lists

Table III-5 ELEMENTS FOUND IN AUTOMOBILE PAINT FINISHES

Paint	Mn	Ba	Zn	Co	Na	Cu	Cr	P	Ratio Ba^{139} to Mn^{56}	Ratio Ba^{131} to Zn^{65}
5	P	P	P	P	P		P		2	1.5
6	P	P	P	X	P	X			4	5
7	P	P	P	D	P	X			10	
8					P	P		P		
9	P			P	P					
10	P	P	P	D	P	X			10	5

P, elements definitely present; X, possible but not definite; D, present but at limit of detection.

the stable elements found in the finishes. The relative amount of each radioactive species present may be determined from the spectral data. Apparent peak-height ratios for Ba^{139} to Mn^{56} and Ba^{131} to Zn^{65} are also given. Since all the samples were irradiated in the same neutron flux, the photopeak heights (radioactivity) for specific radionuclides in one sample may be compared to those found for the same radionuclides in another sample to show the relative abundance of one trace element to another. The intensity of these ratios can be used to identify or selectively separate sample materials.

The same type of procedure was used for soot and soil samples; and, although promising results were obtained, identification of these materials is much more difficult because they contain many more constituents.

Jervis, Perkons, Mackintosh, and Kerr, working with the Crime Detection Laboratory, Royal Canadian Mounted Police, have used activation analysis to determine arsenic in human hair and other tissue to see if this analysis could be used to determine the cause of death in cases of suspected arsenic poisoning.[53] The results to date have been inconclusive. These same workers have made an investigation of the pattern of trace elements in human hair to try to use these patterns for identification purposes. In a case in which activation analysis of a hair was introduced in court,[54] the body of a young woman was found holding a single hair. Samples of hair from the victim and from a male suspect were compared with the hair found in the victim's hand by activation analysis, with the results shown in Table III-6. The difference between the P^{32}-to-Sb^{35} ratio in the two hairs was presented in the Canadian courts as evidence of identity.

Table III-6 NONDESTRUCTIVE ANALYSIS OF SINGLE HAIR IN MURDER INVESTIGATION AT EDMUNDSTON, N. B., CANADA

	β activity*			
	Total, counts/min	P^{32}, counts/min	S^{35}, counts/min	Ratio of P^{32} to S^{35}
Exhibit hair specimen (found in hand of victim)	3540	1783	1757	1.02
Male-suspect hair	2462	1272	1190	1.07
Victim's hair	1908	1276	632	2.02

*β activity after 20 days decay. S^{35}, low-energy component on β-absorption analysis; decay confirmed. P^{32}, high-energy component on β-absorption analysis; decay confirmed.

2. Shipboard Activation Analyzer. A shipboard analyzer[55] performs rapid elemental activation analysis on core samples from the ocean floor on the vessel as they are collected. This aids effective exploration by making it possible for the ship to modify its survey pattern in accordance with the trends indicated by the data. The unit was developed by Dresser Research in conjunction with Lamont Geological Laboratory of Columbia University. The special neutron source is a small, rugged accelerator type neutron generator that emits 10^8 neutrons/sec by the deuteron-tritium reaction. It is contained in a lead and paraffin shield designed to permit insertion of an entire ocean-bottom core sample so that intervals along the core length may be activated without cutting the core. A shielded 3- by 3-in. NaI (Tl) scintillation crystal is used to observe the activity induced in the core, and the gamma spectrum is analyzed in a 400-channel analyzer. Cores are analyzed for silicon, aluminum, magnesium, sodium, and iron contents by neutron activation, and for potassium, uranium, and thorium contents, by the spectra of their natural radioactivities. The activation conditions are: 10 min for irradiation of the core, 1 min for transfer of the core to the detector, and 10 min for measurement of the induced activity. Analyses of standard samples have indicated the following sensitivities in milligrams per cubic centimeter: aluminum and silicon, 0.3; magnesium, 1.6; sodium, 2.2; and iron, 3.8.

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Section

IV

Isotopes and Radiation Technology

Process Radiation Development

One of the exciting applications of the radiant energy from radioisotopes is in the field of process radiation. This includes a broad spectrum of investigations as outlined in the talk by Fowler and Aebersold* and in another talk by Machurek.[1] For example, Yavorsky and Gorin have evaluated beta radiation as a hydrogenation catalyst.[2] Investigators at Battelle Memorial Institute have reported on the influence of radiation on graft copolymerization,[3–5] on radiation-induced nitration of hydrocarbons,[6] and on enhancement of catalytic activity from internal irradiation.[7] Armstrong and Rutherford at North Carolina State College have studied the modification of textile-fiber properties by gamma radiation.[8,9] Other reports cover radiation-induced reactions in swollen polymers,[10] the accelerating effect of additives on radiation-induced graft polymerization,[11] and the mechanism of radiation-induced gelation.[12] The design of beta irradiators is the subject of a report by Weinstock et al. of Radiation Applications Incorporated.[13] A rather comprehensive report from William H. Johnston Laboratories, Inc., deals with basic studies in radiation technology.[14]

Considerable promise is also held for the future of food irradiation, and there is much work going on in this field.[15] Many of the topics mentioned above will be covered in detail in subsequent issues of *Isotopes and Radiation Technology*, with the food problem receiving major attention in the next issue.

Brookhaven National Laboratory (BNL) has been designated by the U. S. Atomic Energy Commission as the Commission's center for studies of the effects of high-intensity radiation. A report on their laboratory, the High Intensity Radiation Development Laboratory (HIRDL), follows.

*See page 1 of this issue.

Radiation Engineering Activities at Brookhaven National Laboratory

By Francis X. Rizzo*

Discussions of the economics of the large-scale applications of radiation in industrial processing invariably are inconclusive because of the lack of engineering experience in the fabrication and handling of megacurie quantities of radiation and the absence of extensive, reliable data on source efficiency. The program of the Division of Isotopes Development[16] at BNL's recently completed HIRDL is directed toward fulfillment of these needs. It is the purpose of this discussion to describe the laboratory and the program.

Facility Design

The facility (Fig. IV-1) includes offices, laboratories, and a hot-cell and canal complex. The complex (Fig. IV-2), designed for the remote handling of 1 megacurie of Co^{60}, is made up of a work preparation cell (13 by 15 by 14 ft high), an experimental irradiation cell (14 by 20 by 14 ft high), and a connecting canal with two side bays. The experimental irradiation cell is a versatile facility designed for large-scale continuous and batch irradiation of gas, liquid, and solid targets. The preparation cell, with its associated special equipment,[17] was designed for use in source decanning, sorting, encapsulating, welding, leak testing, calibrating, and assembling. At present there is approximately 600 kilocuries of Co^{60} on hand.

*Radiation Division, Brookhaven National Laboratory.

Three walls of each of the cells on which equipment controls and viewing windows are located have a thickness of 4.5 ft of 240 lb/cu ft concrete. On the remaining wall and ceiling of each cell, 6.33 ft of ordinary 145 lb/cu ft concrete has been employed. Both cells are equipped with heavy-duty, model 8, manipulators and General Mills mechanical arms, which provide

Each cell is equipped with a remotely operated 24-ton steel-encased lead access door, which is 7 ft 3 in. wide, 8 ft 3 in. high, and 15 in. thick. Pressure-sensitive safety stops on the forward edges of these doors safeguard against injury to personnel or equipment when the doors are in motion. An interlocking safety system controlled by high- and low-level radiation

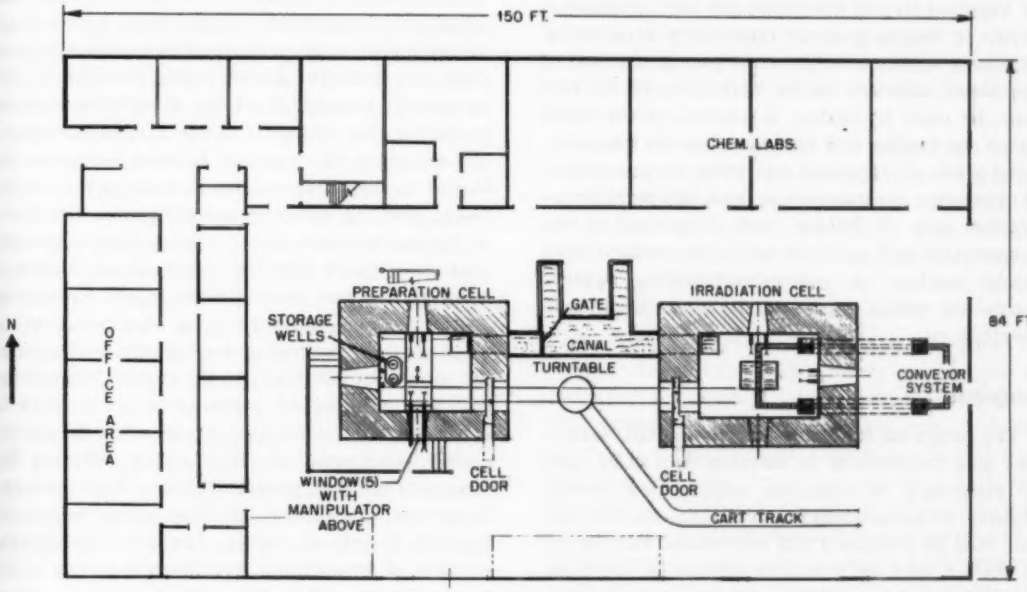

Fig. IV-1 Plan view of High Intensity Radiation Development Laboratory at Brookhaven National Laboratory.

complete cell coverage. Provisions for future installation of periscopes in both cells have been made. The preparation cell, below the floor level, contains two water-cooled isotope storage wells with a capacity of 300 kilocuries of Co^{60}. These wells will be used for the temporary storage of source elements during sorting, calibrating, and encapsulating operations.

Three shielding-glass viewing windows are located in the preparation cell and two, with provision for a third, in the irradiation cell. Each window is 3 by 3.5 ft on the inside, 1.5 by 2.5 ft on the outside, and 54 in. thick. Remotely operated lead shutters on each window serve to minimize the radiation exposure to the window when it is not in use and to protect it against possible damage in the event of an in-cell explosion.

area-monitoring devices regulates the operation of the doors and elevators throughout the facility.

Target material may be moved into and out of the irradiation cell and through the arranged source by means of a conveyor system. The two labyrinth air-duct entranceways that accommodate this system are three-segment U-shaped ducts with two 90° bends. Each duct consists of two vertical shafts, 2.5 by 2.5 by 14 ft deep, and a horizontal section, 2.5 by 6 by 22 ft long, which tunnels under the cell shield. The irradiation cell contains a remotely powered source elevator, which operates inside a water-filled pit that is 5 by 6 by 22 ft deep. Source elements may be assembled into arrays on this elevator and lowered into the water tank when not needed. Heavy equipment and isotope ship-

ping casks may be transported to and from the facility via removable shielding plugs that are located in the ceilings of both cells.

A stainless-steel-lined canal, 45 ft long by 3 ft wide by 16 ft deep, with a 20-ft-deep pit in the center, connects the preparation cell with the irradiation cell. Remotely operated canal lifts move in vertical water-filled shafts that connect the canal to the interior of each cell. At right angles to the canal are two irradiation bays, in which general laboratory irradiation services will be conducted. A gate at the end of the canal adjacent to the work preparation cell may be used to isolate a portion of the canal water for testing and storage of newly encapsulated sources; this will minimize the possibility of spreading contamination from the work preparation cell. A holdup tank connected to the preparation cell collects and holds contaminated liquid wastes. A water-conditioning system treats the water in the canal and in the source elevator pit.

Program

The program for the use of the HIRDL facilities was formulated to develop data to be used in preparing a radiation engineering design manual. Gamma-shielding and dose-distribution data will be compiled and correlated with theory in such a way as to enable engineers who have not had extensive training in radiation physics to arrive at sound irradiator designs. The manual will contain the following 10 chapters: Introduction; Radiation Physics; Computational Procedures; Source Design and Fabrication; Irradiator Design and Fabrication; Health Physics; Source Shipping and Miscellaneous Problems; Economic Considerations; Safety; Appendix.

A major portion of the analytical and experimental effort in compiling such a manual is the development of computational procedures. The calculation problem is similar to that of shielding problems except that, rather than a knowledge of the quality and quantity of radiation escaping from an infinite shield, with perhaps a 20% accuracy, knowledge of the distribution of the radiation flux within a finite target to within a 5% accuracy is required. In most instances the target is less than one mean free path in thickness, and the sources are isotropic. For many of the applications under consideration, specifications require that the ratio of the maximum dose at any two points in the target not exceed 1.25. Various computational methods may be applied—the method of successive scatterings, the moments method, and the Monte Carlo method. The method of successive scatterings is limited and, in general, inapplicable. An appreciable background of calculations utilizing the moments method, primarily those of Spencer and Goldstein, does exist, but these data are applicable only to infinite media. The only appropriate method of importance for finite systems is the Monte Carlo method. Monte Carlo techniques are at present being applied for handling the stated problem. The cooperation of the Bureau of

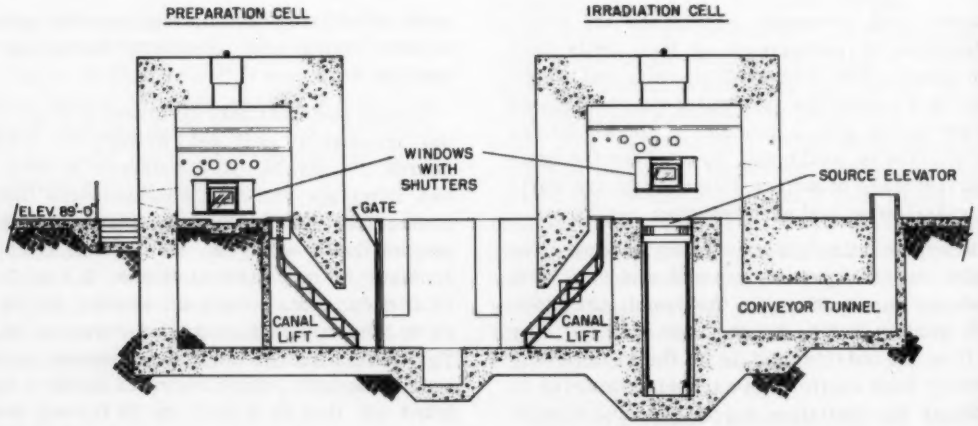

Fig. IV-2 Section view of cell-canal complex of the High Intensity Radiation Development Laboratory at Brookhaven National Laboratory.

Standards and Yale University has been sought, and collaboration with these groups is under way. These techniques will ultimately make possible the handling of a wide variety of dose-distribution problems.

There has been a continuing effort at BNL to obtain experimental dose-distribution data. Work with large-scale sources of low activity levels has made considerable data available for point, cylindrical, and slab sources, principally with homogeneous, unit density, infinite, semi-infinite, and finite targets.[18,19] Simultaneously, a program for determining experimental buildup factors for these geometries has been under way.[20] At present, experimental boundary buildup factors are being formulated for one, two, and three boundary cases.

The problem of the scattering of gamma radiation through air ducts in concrete shields has been investigated. The study was made with large, rectangular, two- and three-segment ducts with 90° turns. Equations based upon experimental data involving the duct parameters have been formulated.[21,22] Although these studies have not as yet led to analytical techniques of duct design, efforts along these lines are continuing. Recent experimental determinations of the spectral distribution of multiply scattered gamma rays inside air-concrete ducts may facilitate the applications of an analytical solution to the problem.

The experimental measurement of scattered photon spectra is of considerable interest. Using the theoretical and experimental response matrix of a scintillation spectrometer, the observed spectrum is converted to the actual spectrum in the target. This approach employs the use of small cylindrical scintillators ($\frac{1}{2}$ in. in diameter by $\frac{1}{2}$ in. high plastic and sodium iodide crystals) and an unfolding technique that tests the corrections to the observed spectrum and yields the most probable true spectrum. An IBM-7094 spectrum-unfolding FORTRAN program, which yields a spectrum with a probable accuracy of 5%, is presently in use. Measurements based upon these techniques will be useful in the study of the effect of boundaries and inhomogeneities and on multiple-scattering gamma-ray problems, which thus far have been solved stochastically by the Monte Carlo techniques.

Analytical and experimental efforts for developing generalized methods for irradiator design are in progress. A considerable amount of data has been generated aimed at evaluating irradiator design parameters such as source energy and overlap, source-to-target air gap, source-element design and standardization, encapsulation, testing, and safety.[23] Design parameters have been studied for Co^{60}, Na^{24}, and Cs^{137} slab irradiator systems.[24] The effect of heterogeneous targets on design parameters has been considered in these studies.

Analytical methods are being developed to treat a limited number of standard cases of combinations of moving sources and/or moving targets by equating them mathematically to static cases.

In general, these design and analytical efforts are aimed at optimizing the efficiency of gamma-ray and gamma-energy utilization. The efforts are aimed not only at developing a particular design or obtaining a particular piece of information but also toward gaining a better understanding of the fundamentals involved.

The intense irradiation of shielding glass by gamma rays has been known to result in catastrophic internal electrical discharge. At BNL studies of the problem of gamma-induced electrical breakdown in cerium-protected lead shielding glasses from an experimental and theoretical viewpoint are being conducted, with effort concentrated toward balancing both fundamental and practical viewpoints. A theoretical calculation involving classical electromagnetic theory and a theory of separation of charge has been carried out to determine the buildup of electrical potential inside typical shielding glasses as a result of irradiation with gamma rays. The experimental program has been designed to test the predictions of the above theory and to correlate the breakdown process with other phenomena such as cerium color center formation and fading, the buildup of temperature gradients, and the time duration and pattern of the discharge.

Preliminary experiments indicate a correlation between the cerium content of the glass and the total dose at which controlled discharge will occur. Glasses with the highest cerium content experience electrical breakdown more readily and at lower total doses than those with low cerium content.

Concomitant with the experimental program described thus far, a sizable effort has been expended toward the development, testing, and evaluation of new high-level dosimetry systems. Among the systems that have been developed at

BNL are two silicon solar-cell dosimeters[25,26] and two modified "Fricke" dosimeters.[27] The two silicon solar-cell dosimeters are the *n* on *p* type dose rate meter, with a range from 10^2 to 10^9 rads/hr, and the *p* on *n* type integrating dosimeter, with a total dose range from 10^5 to 10^8 rads. The ferrous-ferric sulfate chemical dosimeters studied at BNL are modifications of the conventional Fricke dosimeter and may be referred to as the "aerated" and the "deaerated" Fricke dosimeters. Both methods were employed to extend the range of the Fricke dosimeter from 10^4 rads to well beyond 10^6 rads.

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Section

V

Isotopes and Radiation Technology

Source Development

The decision to separate this section on Source Development from the section on Isotopic Power Development (Sec. VI) was purely arbitrary. Section VI is oriented toward the large-scale applications, whereas this section emphasizes the research and development activities. Overlap, of course, is inevitable.

Glasses

By R. E. Lewis

Recent trends are toward preparing radioisotopic sources as insoluble compounds or in insoluble matrices to decrease the contamination hazard if the primary encapsulation is destroyed. Low leach rates and high abrasion resistance make glass attractive as a radiation source material. This is especially true for radioactive monovalent ions, such as Cs^{137}, whose salts are very soluble. Much of the recent work at Oak Ridge National Laboratory (ORNL) has been directed toward preparing glasses from the fission products, with strong emphasis on the preparation of cesium glasses and evaluation of their properties. Glasses prepared at ORNL include aluminosilicate,[1] silicate,[2–6] and borosilicate,[7,8] with major emphasis on incorporating cesium into these glasses for use as radioactive sources.

Aluminosilicate Glasses

Aluminosilicate glasses are prepared by adsorbing a radioactive cation on a synthetic aluminosilicate ion exchanger (Decalso), mixing with a flux, and fusing at a high temperature. Although especially good for making cesium sources, any radioactive cation can be adsorbed on Decalso. The ion exchange capacity of Decalso is ~2.0 meq/g; however, the amount of a carrier-free tracer which can be adsorbed on the ion exchanger depends on the distribution coefficient of that ion, which in turn depends on the salts present, their concentrations, and the selectivity of the ion exchanger for that ion rather than entirely on the capacity of the exchanger.

Adsorption Properties of Decalso. Cations that have been found to be strongly adsorbed on Decalso are cesium, rubidium, calcium, strontium, barium, scandium, titanium, chromium, manganese, iron, cobalt, nickel, copper, zinc, yttrium, zirconium, Ru(III), silver, indium, cadmium, the rare earths, gold, mercury, thallium, lead, americium, uranium, and neptunium. Capacity is maximum at the *p*H of incipient hydroxide precipitation. Anions, in general, do not adsorb on Decalso, although $P^{32}O_4^{3-}$ adsorbs on the zinc form of Decalso at *p*H 10 with a distribution coefficient of ~30.

The Decalso used as a source matrix is first ground to ~200 mesh, and the desired amount of material is contacted with the desired radioisotope solution for 1 to 2 hr. Exchange is complete for tracer concentrations in 30 min, but greater concentrations of cations require up to 2 hr for 90 to 95% adsorption and 24 hr for ~99% adsorption.

Glass-Preparation Studies. Decalso (7.5% Na_2O, 15.9% Al_2O_3, 49.5% SiO_2, and 27.1% combined water) melts at 1450 to 1500°C to a glass. Compounds tested as fluxes to lower the melting range and viscosity of the glass include B_2O_3, CaF_2, and CaO; however, only CaO gave a glass resistant to leaching. The melting points of Decalso-CaO mixtures are shown in Table V-1. Since 8 to 12% CaO gave the lowest melting glass, 10% CaO was used in the glass prepared for leaching tests. The composition of this glass after firing is 8.8% Na_2O, 18.7% Al_2O_3, 57.7% SiO_2, and 13.8% CaO. The glass is

Table V-1 MELTING POINTS OF DECALSO-CaO MIXTURES

Composition, wt.%		Melting point, °C
Decalso	CaO	
100	0	1400
96.5	3.5	1240
92.5	7.5	1180
90.0	10.0	1160
88.5	11.5	1180
85.0	15.0	1210
80.0	20.0	1230
75.0	25.0	1250
70.0	30.0	1280
67.0	33.0	1290
56.5	43.5	1320
53.0	47.0	1400

harder than ordinary plate glass and very resistant to thermal and mechanical shock.

Leaching Tests. In a 400-hr leach of the glass at room temperature, two to five times as much activity was removed as in a 68-hr leach (Table V-2). The glass was resistant to all acids, bases, and salts tested except HF. The tests were made on 0.1-mc sources of Cs^{137}, Sr^{89}, Tl^{204}, and Ce^{144}.

Source Preparation. Decalso can be used to prepare glazes that have high abrasion resistance and low solubility, yet are thin enough to be useful as sources of soft beta or alpha radiation.

Table V-2 LEACHABILITY OF DECALSO GLASS SOURCES

Leaching solution	Isotope leached from 1-cm^2, 0.1 mc source in 68 hr at 25°C, %			
	Cs^{137}*	Tl^{204}	Sr^{89}	Ce^{144}
Distilled water	$<1 \times 10^{-5}$	$<1 \times 10^{-5}$	$<1 \times 10^{-5}$	$<1 \times 10^{-5}$
Process water	$<1 \times 10^{-5}$	4×10^{-5}	1×10^{-5}	1×10^{-5}
8*M* HNO_3	1×10^{-2}	3×10^{-2}	2×10^{-2}	1×10^{-2}
6*M* HCl	3×10^{-2}	3×10^{-3}	3×10^{-3}	3×10^{-3}
12*M* H_2SO_4	1×10^{-2}	6×10^{-2}	2×10^{-2}	1×10^{-3}
25% HF	7.0	10.0	3.0	4.0
6*M* NaOH	2×10^{-2}	1×10^{-3}	3×10^{-2}	1×10^{-2}
4*M* NH_4OH	1×10^{-5}	2×10^{-4}	2×10^{-3}	1×10^{-4}
4*M* $NaNO_3$	1×10^{-3}	1×10^{-2}	4×10^{-2}	1×10^{-4}
4*M* NH_4NO_3	1×10^{-3}	1×10^{-2}	4×10^{-2}	5×10^{-5}
1*M* NH_4F	5×10^{-3}	5×10^{-3}		
Simulated sea water	1×10^{-4}	1×10^{-3}	1×10^{-3}	1×10^{-4}

*Loading equivalent to 6 curies of Cs^{137} per gram.

Ninety-nine percent of the activity of 7.4 mc of Sr^{89} was adsorbed on 0.5 g of Decalso in 24 hr. The Decalso was mixed with 10% CaO and fused at 1300°C in an Alundum cup to give a glass layer 0.01 in. thick. There was no detectable loss of Sr^{89} on ignition. The source surface smeared 10^3 counts/min, and, after 2 days' equilibration with 2*M* HNO_3, 8×10^3 counts/min, indicating that etching does not decrease the count. The observed radiation output was 3.1 r/hr at 20 cm; the calculated value was 3.5 r/hr, assuming no self-absorption in the source.

Alpha sources can be prepared from heavy radioisotope Am^{241}, $U^{233}O_2$, or $Np^{237}O_2$ ions by adsorption on Decalso. Thin films of glass (glazes), 0.3 to 5.0 μ thick, are fused to ceramic or stainless steel. Finely ground glass mix (75% Decalso + 25% B_2O_3, m.p. 1000°C, is used with stainless steel), in benzene containing 1 to 2% Canada balsam, is spread on the backing material, and, after drying, is fused at 1000°C to form a 0.1-mg/cm^2 layer. The efficiency of the alpha sources as a function of the glaze thickness is shown in Table V-3.

Table V-3 EFFICIENCY OF Am^{241} ALPHA SOURCES OF VARIOUS THICKNESSES

(1 mg of Decalso Yields a Total of 6885 dis/min α)

Decalso in glass, mg/20 cm^2	Calculated glass thickness, μ	Measured alpha		Efficiency, %
		dis/min	dis/(min)(mg)	
1.20	0.3	8,088	6735	97.8
2.20	0.6	14,773	6714	97.5
4.60	1.3	30,200	6564	95.3
4.90	1.5	34,280	6996	101.6

Abrasion Resistance. Table V-4 shows the abrasion-resistance properties of the Decalso-B_2O_3 sources.

Granular Sources. Small sources capable of giving a high radiation dose can be prepared by absorbing a beta emitter (Pm^{147}, Sr^{90}, Cs^{137}, or Ce^{144}) on 325-mesh Decalso and firing to about 1000°C for 1 hr. At this temperature, although the Decalso particles do not fuse, the grains become glassy and the radioactivity is not leached. Since the particles are smaller than 325 mesh (44 μ), there is little beta self-absorption, especially with Sr^{90} and Ce^{144}. Since this type of source is easily prepared, it would

be useful for short-half-life fission products as well as for the longer-lived isotopes.

If complete beta absorption is assumed, the maximum dose rate may be calculated in rads per hour per gram of absorber per curie for Pm^{147}, Sr^{90}, Ce^{144}, and Cs^{137}, where T_0 is the maximum beta energy:

$Pm^{147} \rightarrow Nd^{147} + \beta$ T_0 = 0.23 Mev per disintegration
$Sr^{90} \rightarrow Y^{90} + \beta$ T_0 = 0.54 Mev per disintegration
$Y^{90} \rightarrow Zr^{90} + \beta$ T_0 = 2.27 Mev per disintegration (total, 2.81 Mev per disintegration)
$Ce^{144} \rightarrow Pr^{144} + \beta$ T_0 = 0.30 Mev per disintegration
$Pr^{144} \rightarrow Nd^{144} + \beta$ T_0 = 2.98 Mev per disintegration (total, 3.28 Mev per disintegration)
$Cs^{137} \rightarrow Ba^{137} + \beta$ T_0 = 0.52 Mev per disintegration

Calculated dose rates when 100% absorption of the activity is assumed are shown in Table V-5.

Silicate Glasses

Cesium silicate glasses were prepared from SiO_2, carbonates of the divalent metals, and cesium carbonate; the Cs_2CO_3 was prepared from cesium alums by the cesium tetraoxalate process. Silicate glasses have the general formula $xR_2O \cdot yRO \cdot zSiO_2$, where $x + y = 1$, and z varies from 2.5 to 4.0. The R_2O component can be Na_2O, K_2O, or, in the case of cesium glasses, Cs_2O. The function of the R_2O group is to decrease the viscosity of the glass; however, the proportion of R_2O should not be greater than 0.8, or the leaching rate of the glass will be excessive. The function of the RO group is to decrease the leaching rate and to improve the annealing properties of the glass. The SiO_2 is the major glass-forming constituent and should be present to the extent of 2.0 to 4.0 moles per mole of the basic oxides (R_2O, RO). Glasses containing less

Table V-4 ABRASION RESISTANCE OF DECALSO-B_2O_3 GLAZE

Source	Activity per cm^2	Thickness	Smear material	Smear, dis/(min)(cm^2)
Cs^{137}	1 μc	1 mm	Smear paper	<10
			Crocus cloth	<10
Cs^{137}	100 μc	1 mm	Smear paper	<10
			Crocus cloth	100
Sr^{90}	1 mc	1 mm	Smear paper	2000
			Crocus cloth	4000
Sr^{90}	7 mc	1 mm	Smear paper	4000
			Crocus cloth	4000
Tl^{204}	100 μc	1 mm	Smear paper	<10
			Crocus cloth	100
Ce^{144}	100 μc	1 mm	Smear paper	<10
			Crocus cloth	100
U^{233}	10^6 dis/min	0.1 μ	Smear paper, dry	<5
			Smear paper, wet	5
			Crocus cloth, dry	120
			Crocus cloth, wet	240
			Carborundum paper (000)	600

Table V-5 CALCULATED DOSE RATES FOR GRANULAR SOURCES

(Assumptions: 1 g of Decalso, 1 curie of Activity, and 100% Absorption of Activity)

Isotope	Dose rate,* rads/(hr)(curie)	Approximate specific activity, curies/g	Decalso capacity, curies/g	Maximum source strength, rads/(hr)(g of Decalso)
Pm^{147}	1.6×10^5	400	35	5.6×10^6
Sr^{90}	1.9×10^6	80	7.2	1.4×10^7
Ce^{144}	2.3×10^6	300	27	6.2×10^7
Cs^{137}	3.6×10^5	33	8.7	3.1×10^6

* $\frac{\text{rads}}{(\text{hr})(\text{curie})} = \frac{(\text{Mev})}{(3\ \text{dis})} \frac{(2.2 \times 10^{12}\ \text{dis})}{(\text{min})(\text{curie})} \frac{(60\ \text{min})}{(\text{hr})} \frac{(1.6 \times 10^{-6}\ \text{ergs})}{(\text{Mev})} \frac{(\text{rad})}{(100\ \text{ergs})}$.

Table V-6 EFFECT OF RO GROUP ON CESIUM LEACHING RATE, DENSITY, AND MELTING RANGE

Empirical formula	Cesium content		Density, g/cm³	Cesium leaching rate, mg/(cm²)(day)	Melting range, °C
	Wt.%	Curies/g of glass			
$0.15Li_2O \cdot 0.55Cs_2O \cdot 0.3CaO \cdot 2.7SiO_2$	41.5	41.0	3.00	1.7	1300 to 1350
$0.15Li_2O \cdot 0.55Cs_2O \cdot 0.3ZnO \cdot 2.7SiO_2$	40.5	43.1	3.22	2.4	1250 to 1300
$0.15Li_2O \cdot 0.55Cs_2O \cdot 0.3BaO \cdot 2.7SiO_2$	38.0	40.3	3.21	2.0	1200 to 1250
$0.15Li_2O \cdot 0.55Cs_2O \cdot 0.3CdO \cdot 2.7SiO_2$	38.8	41.8	3.26	2.7	1100 to 1150
$0.15Li_2O \cdot 0.55Cs_2O \cdot 0.3PbO \cdot 2.7SiO_2$	36.0	44.0	3.78	4.7	1050 to 1100
$0.15Li_2O \cdot 0.55Cs_2O \cdot 0.3SrO \cdot 2.7SiO_2$	39.8	41.5	3.17	2.0	1300 to 1350

than 2.0 moles of SiO_2 tend to crystallize, whereas those containing greater than 4 moles of SiO_2 are too viscous to flow well at 1200 to 1300°C.

Table V-6 shows the effect of various divalent oxides on the leaching rate, melting range, and density of the glass. Although there is little difference in the leaching rates of glasses of similar compositions, some of the oxides have disadvantages. Zinc oxide dissolves slowly in the glass melt and has a strong tendency to cause devitrification of the glass. Barium oxide glasses low in SiO_2 also have a strong tendency to devitrify after long heating at 800°C, but this is not true with >3.0 SiO_2. Cadmium oxide volatilizes from the melt at 1150°C or higher, especially if a stream of air is passing over the melt. Lead glasses must be made in an oxidizing atmosphere to prevent formation of lead metal. Calcium and strontium oxides give good cesium glasses, melting at 1300 to 1350°C, whereas barium oxide glasses melt lower, at 1200 to 1250°C. All the glasses in Table V-6 contain 35 to 45 wt.% cesium. Table V-7 shows the effect of the R_2O:RO ratio on both the leaching rate in water at 25°C and the volatilization of cesium at 1200°C in the Cs_2O-CaO-SiO_2 system.

Table V-7 EFFECT OF THE R_2O:RO RATIO ON THE LEACHING RATE AND VOLATILIZATION OF CESIUM FROM $Cs_2O \cdot CaO$ GLASSES

Glass	Cs_2O volatilization rate at 1200°C, mg/(cm²)(hr)	Cesium leaching rate, mg/(cm²)(day)
$0.5Cs_2O \cdot 0.5CaO \cdot 2.7SiO_2$	0.5	<1.0
$0.55Cs_2O \cdot 0.45CaO \cdot 2.7SiO_2$	0.7	<1.0
$0.60Cs_2O \cdot 0.4CaO \cdot 2.7SiO_2$	1.2	1.0
$0.65Cs_2O \cdot 0.35CaO \cdot 2.7SiO_2$	1.5	1.3
$0.70Cs_2O \cdot 0.30CaO \cdot 2.7SiO_2$	2.0	1.7
$0.75Cs_2O \cdot 0.25CaO \cdot 2.7SiO_2$	5.0	5.0
$1.00Cs_2O \cdot 2.7SiO_2$	9.0	Very soluble

Table V-8 EFFECT OF SiO_2 ON PROPERTIES OF $Li_2O \cdot Cs_2O \cdot BaO \cdot SiO_2$ GLASSES

Glass	Cs_2O volatilization at 1200°C, mg/(cm²)(hr)	Cesium leaching rate in H_2O, mg/(cm²)(day)	Remarks
$0.15Li_2O \cdot 0.55Cs_2O \cdot 0.3BaO \cdot 2.0SiO_2$	10.0	10.0	Glass tends to be devitrified
$0.15Li_2O \cdot 0.55Cs_2O \cdot 0.3BaO \cdot 2.5SiO_2$	4.0	5.0	Glass tends to be devitrified
$0.15Li_2O \cdot 0.55Cs_2O \cdot 0.3BaO \cdot 2.7SiO_2$	2.0	2.0	Glass tends to be devitrified on long heating
$0.15Li_2O \cdot 0.55Cs_2O \cdot 0.3BaO \cdot 3.0SiO_2$	1.5	1.1	Good glass
$0.15Li_2O \cdot 0.55Cs_2O \cdot 0.3BaO \cdot 3.5SiO_2$	1.0	0.5	Good glass
$0.15Li_2O \cdot 0.55Cs_2O \cdot 0.3BaO \cdot 5.0SiO_2$	0.4	0.3	Viscous glass at 1200°C
$0.15Li_2O \cdot 0.55Cs_2O \cdot 0.3BaO \cdot 10.0SiO_2$	0.2	0.1	Very viscous glass at 1200°C

Generally, cesium leaching and volatilization rates from silicate glasses are low. However, glasses with a high calcium oxide content have a lower volatilization rate at 1200°C and a lower leaching rate than those containing less divalent oxide. The SiO_2 content also has a great influence on the leaching and volatilization rates of cesium oxide from the glass. Table V-8 shows that an increase in the SiO_2 content lowers the leaching and volatilization rates of cesium at 1200°C from the $Li_2O \cdot Cs_2O \cdot BaO \cdot SiO_2$ system.

A good cesium glass for molding glass bars is $0.8Cs_2O \cdot 0.2BaO \cdot 3.5SiO_2$. This glass contains 45.7% cesium and has a density of 3.1 g/cm^3, giving a Cs^{137} activity concentration of 47.4 curies/cm^3. The leaching rate is <2 mg/(cm^2)(day), and the volatilization rate, <2 mg/(cm^2)(hr) at 1200°C. The glass does not have a tendency to devitrify and can be annealed at 500°C. This glass would be useful for molding cesium rods or bars for a radiation source, or right cylinders for heat sources.

Borosilicate Glasses

Cesium borosilicate glass, $Cs_2O \cdot B_2O_3 \cdot 4.2SiO_2$, has been prepared at ORNL by precipitating cesium tetraphenylboron with 240-mesh SiO_2, calcining at 700°C to form $Cs_2O \cdot B_2O_3 \cdot 4.2SiO_2$, and melting at 1200 to 1300°C. The glass contains 44 wt.% cesium and has a density of 3.1 g/cm^3. At 1200°C it volatilizes at a rate of 35 mg of $Cs_2O \cdot B_2O_3$ per square centimeter per hour. The cesium leaching rate of such a glass is 0.2 mg/(cm^2)(day). Cesium borosilicate glasses are difficult to anneal and have a tendency to crystallize if cooled too slowly in the temperature range 1100 to 1200°C.

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Section

VI

Isotopes and Radiation Technology

Isotopic Power Development

The heat energy generated by the absorption of radiations from radioisotopes during decay may be partially converted to electricity and thus used as a power source. Such thermoelectric generators are suitable for both terrestrial use in remote areas and space use.[1] The generators for remote-area terrestrial use are discussed here, which, in the present state of development, are based largely on fission-product radioisotopes. Most of the current space applications use alpha-emitting isotopes[2] and are not discussed here. Properties of radioisotopes that are of interest for making power sources are reviewed in Refs. 3 and 4. Continuing work on fission products is reported regularly.[5]

Radioisotope Power Sources

In 1957 Hittman[6] presented availability and cost studies that showed the feasibility of specialized low-power-output applications for radioisotopes. Subsequent research carried out under contract with the U. S. Atomic Energy Commission, Division of Isotopes Development, in cooperation with other government agencies has resulted in development of radioisotope-fueled generators.

A 5-watt thermoelectric generator (Fig. VI-1) fueled with 17,500 curies of Sr^{90} and an automatic nuclear-powered meteorological data-transmitting radio station for unattended service were designed, manufactured, and tested by the Martin Co. for the Division of Isotopes Development and the U. S. Weather Bureau. After extensive testing the system was installed on Axel Heiberg Island in the Arctic in 1961 and is still operating satisfactorily.[7,8] The system automatically measures air temperature, barometric pressure, wind direction, and two wind-speed averages. Once every 3 hr it transmits the data in binary digital form to a manned receiving station, where it is recorded on a two-channel

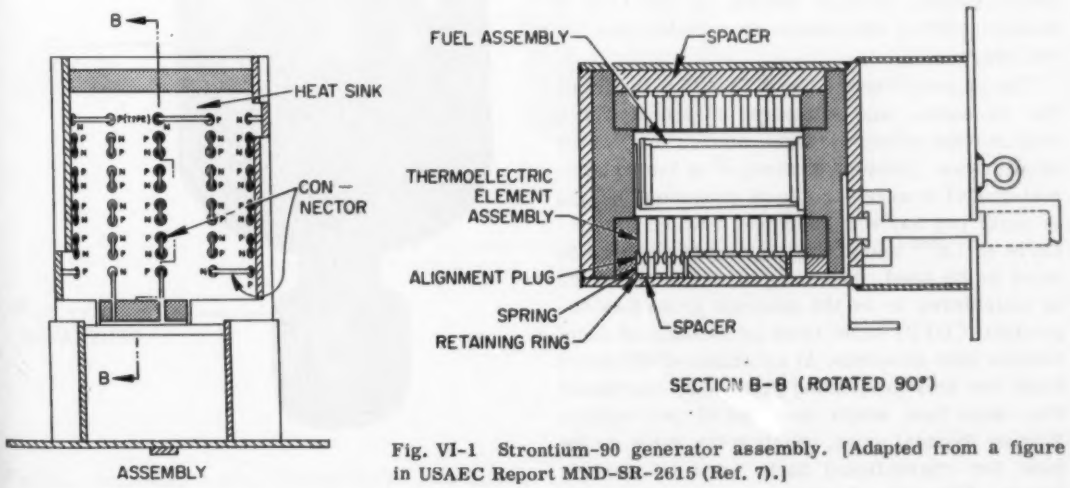

Fig. VI-1 Strontium-90 generator assembly. [Adapted from a figure in USAEC Report MND-SR-2615 (Ref. 7).]

recorder. Temperature, wind direction, and barometric pressure are read at the instant of transmission. Wind speed is averaged over an 8- and a 1-min interval immediately before transmission. The generator and data-telemetry package are in an insulated cavity 8 ft below the ground surface. Although the permafrost temperature varies from −10 to +30°F, the 95 watts of waste heat manufactured keeps the cavity between 40 and 80°F.

The strontium is in the form of strontium titanate, whose solubility in 0.1*N* acid or base at 120°F in 750 hr was shown to be less than 5 ppm. Tests in both fresh and sea water indicated that more than 900 years would be required to dissolve the pellets tested. Strontium titanate[9] melts at 1910°C, whereas 600°C is the maximum expected fuel temperature in a $Sr^{90}TiO_3$ thermoelectric generator. The Hastelloy C capsule used for the fuel is not pitted in sea water and is corroded at a rate of 0.0001 in. per year.

A Sr^{90} titanate-fueled generator (SNAP-7A, Fig. VI-2) was also developed for a U. S. Coast Guard light buoy.[10] The generator supplies a nominal 10 watts of electric power at the end of the 10-year design life with an operating voltage of about 5 volts and is designed for operation without maintenance or attendance for two years. The fuel is encapsulated in Hastelloy C and is shielded with depleted uranium. The SNAP-7D 60-watt generator[11] is also fueled with Sr^{90} titanate, which is sealed in Hastelloy C. It is designed to provide electric power for the U. S. Navy floating weather station in the Gulf of Mexico without attendance or maintenance for two years.

The Chance Vought Corp. issued a report on the economic and technical feasibility of a radioisotope-powered heat source for water desalination (Project Artesia).[12] A laboratory-test-model source capable of producing 250 gal of water per day was designed, in which 1 megacurie of Ce^{144} and a six-stage flash evaporator were to be used. The most economical source is considered to be the calcined gross fission-product (CGFP) waste from processing of spent reactor fuel elements. At a distance of 400 miles from the fuel-processing plant, they calculated that such fuel would cost $0.55 per million British thermal units, which is the same as the cost for conventional fuels used to produce steam. The use of various separated fission products was also investigated.

Cesium-137 as a power source has been investigated by the Royal Research Corporation, with special emphasis on underwater applications.[13] A complex borosilicate glass, containing 43 wt.% cesium, was found to have a very low solubility and to be suitable as the heat source. After numerous safety tests, a prototype thermoelectric generator was designed to convert the decay heat from 27,000 curies of Cs^{137} (124 thermal watts) to 5 watts of electricity (Fig. VI-3). On the basis of this preliminary work, Royal Research Corporation and Oak Ridge National Laboratory (ORNL) fabricated a source for an underwater seismograph device.

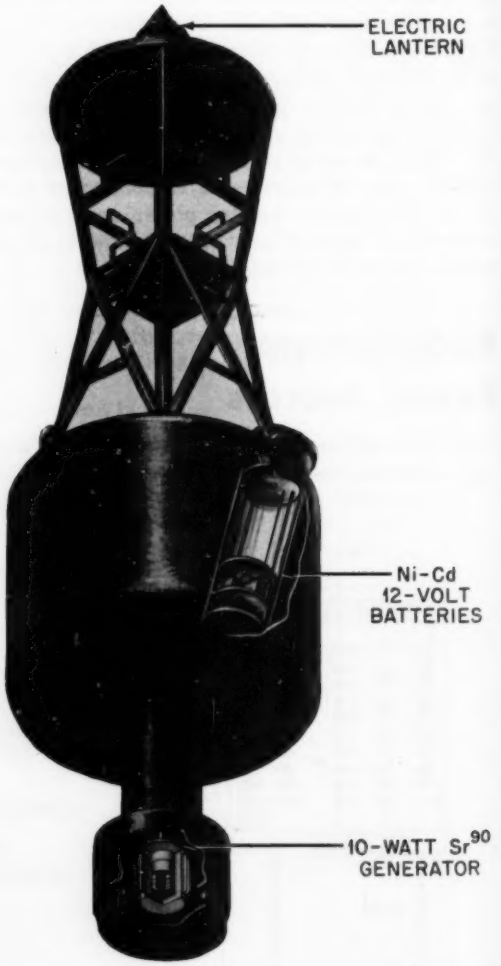

Fig. VI-2 Thermoelectric generator system[10] mounted in standard U. S. Coast Guard flashing light buoy and fueled with Sr^{90}.

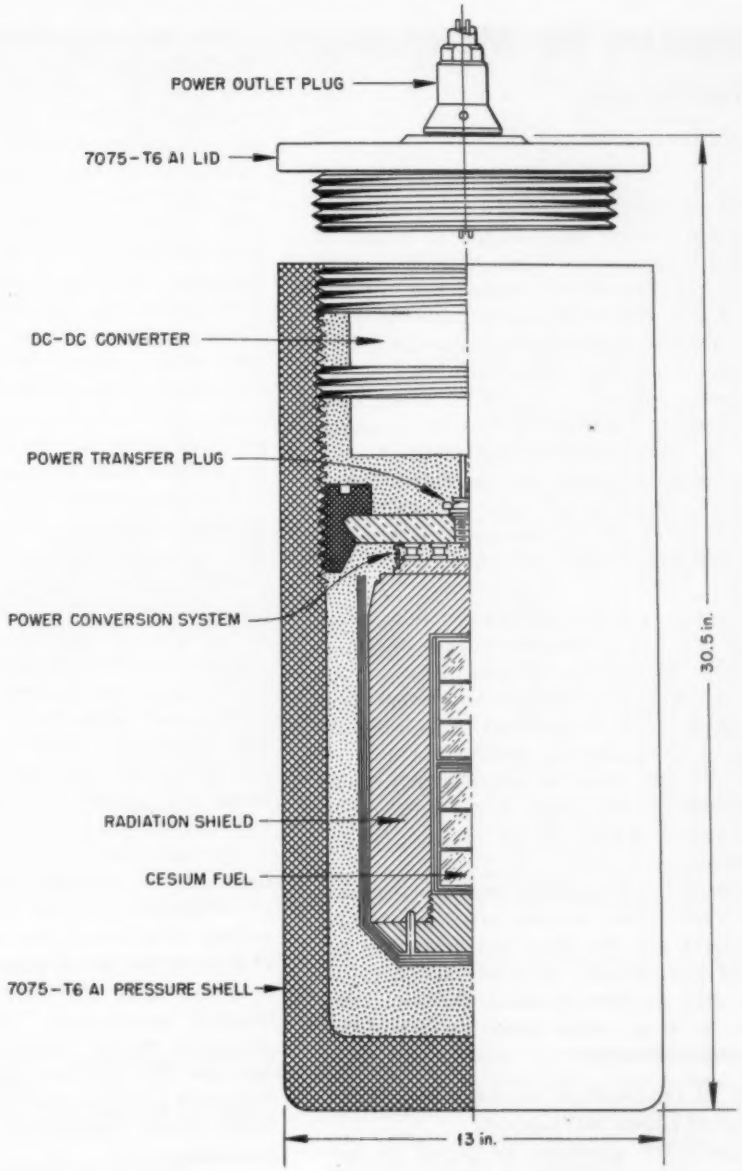

Fig. VI-3 Five-watt thermoelectric generator powered with Cs^{137}. [Adapted from a figure in Report RRC-0103 (Ref. 13).]

Calorimetry for Measuring Radioactivity

By J. C. Posey

The use of calorimetry at the ORNL Isotopes Development Center was started as a part of the program to produce large radioactive heat sources. A calorimetric method was desired since it would give direct measurements of the rates of heat production and thus would eliminate errors associated with uncertain knowledge of the isotope decay schemes. It is the only feasible method of measuring directly the radioactivity of large samples that produce only alpha or beta radiation. The dependability of calorimetric methods and the fact that they have generally greater accuracy than competing counting procedures have led to their increasing use.

Calorimetry as applied to radioactive sources differs in one basic respect from conventional calorimetry: the quantity measured is power, or rate of heat production, and is measured in such units as watts or calories per second; in conventional calorimetry a quantity of heat is measured in units such as watt-hours or calories. Because of this, most commercial calorimeters cannot be used for the assay of radioactive sources.

A number of calorimeters for assaying the power of radioactive sources are described in Refs. 14 and 15; the first was described by Rutherford and Barnes[16] in 1904. These calorimeters vary greatly in detail. However, the operation is in all cases based on one of the following measurements:

1. Rate of temperature increase of a mass of material of known heat capacity
2. Temperature gradient produced by the conduction of heat through a path of known properties
3. Temperature increase of a constant stream of cooling fluid
4. Rate of vaporization of a liquid

Most of the calorimetric work in the ORNL Isotopes Development Center has been done with simple calorimeters developed in its own laboratories; these are of type No. 2 mentioned above. They occupy a minimum of hot-cell space and are easy to operate, resistant to damage, and low in cost.

The basic parts of these calorimeters are shown in Fig. VI-4. The upper part is a metal cup mounted on a metal shaft, at the base of which is a heat sink cooled by a stream of water at constant temperature. The heat produced by radioactive decay of the source is absorbed by the cup and conducted down the shaft to the heat sink. At steady state the temperature difference between the top and the base of the shaft is nearly proportional to the rate at which heat is produced by the radioactive source. The temperature difference is measured by a thermopile with junctions attached to the surface of the shaft. Copper wires are attached to the thermopile at the hot end of the shaft. Extraneous heat loss is minimized by enclosing the cup and the shaft in an inverted Dewar flask.

The construction materials were selected to maintain nearly exact proportionality between the thermopile output and the thermal power of the source. Iron-constantan couples were chosen for the thermopile because their sensitivity (millivolts per degrees centigrade) varies only slightly within the range of temperatures measured. Copper was selected for the shaft because its thermal conductivity is relatively constant. The departures from constancy of thermopile sensitivity and thermal conductivity which do occur are opposite in effect to the small loss of heat through the Dewar flask.

The calorimeters are calibrated before use by means of an electric heater placed in the cup in substitution for the radioactive source. The voltage drop across the heater, the heater current, and the thermopile output are measured with precision electrical equipment of commercial manufacture. These electrical measurements during calibration and those during use are the only instances during which a high degree of precision must be attained. Comparatively loose tolerances are permissible in fabrication.

The design of a calorimeter of this type involves a suitable compromise between accuracy and speed of operation. The approach to steady state is described approximately by

$$\theta = \frac{C}{U} \ln \frac{\Delta T_{ss}}{\Delta T_{ss} - \Delta T}$$

where θ = elapsed time
C = heat capacity of the sample cup and upper part of the Dewar flask (the primary assumption is that these are at the same temperature)

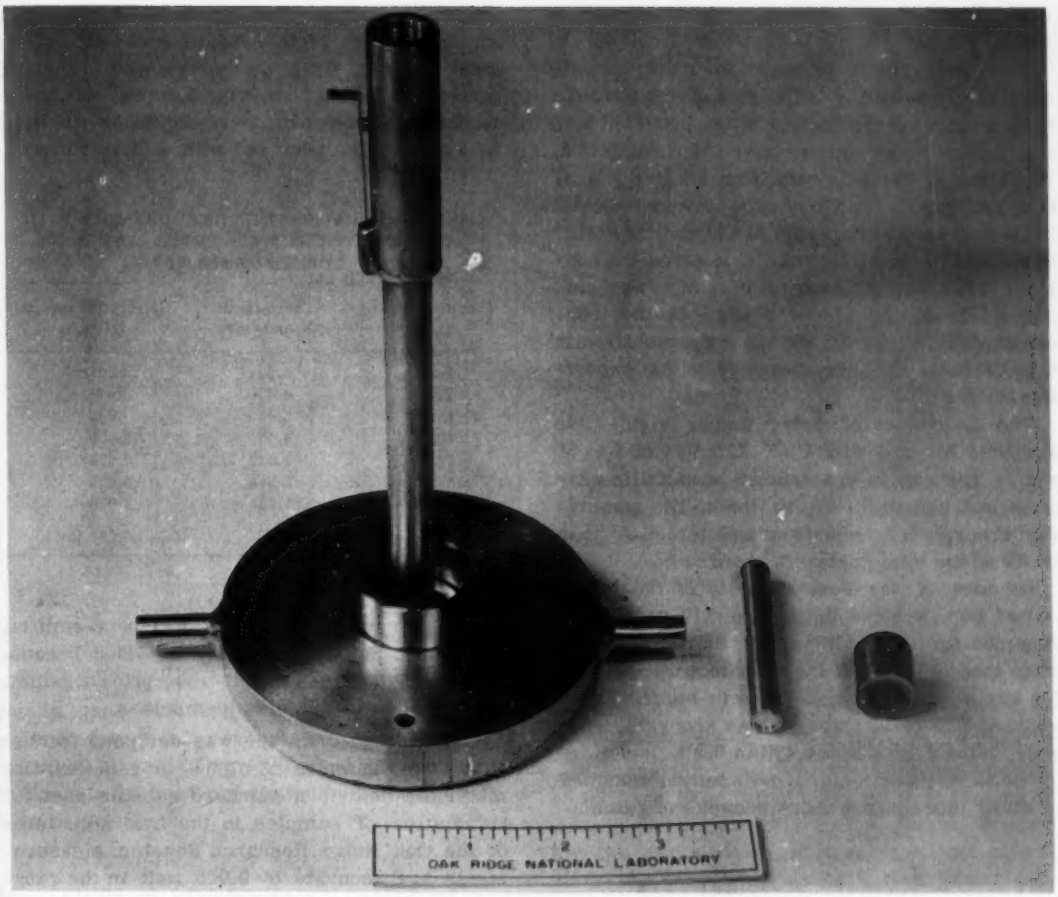

Fig. VI-4 Basic parts of calorimeter used for measuring rates of heat production by radioisotope sources.

ΔT = difference in temperature between ends of shaft

ΔT_{ss} = difference in temperature at steady state

U = thermal conductivity of shaft

The time required for ΔT to reach a given fraction of ΔT_{ss} is thus inversely proportional to the thermal conductivity of the shaft. However, ΔT_{ss} is inversely proportional to U. Consequently, since the error caused by fluctuations in cooling-water temperature and by limits of precision of the measurement of the thermopile output are approximately inversely proportional to ΔT_{ss}, the increase in speed of operation is accompanied by a loss of precision. Furthermore, if U is increased until the resistances to heat flow from the sample to the cup and from the cup to the Dewar flask are significant compared to the shaft resistance, the time required will not decline as rapidly as predicted by the equation.

Four of the calorimeters that have been constructed are shown in Fig. VI-5, minus the Dewar flasks and thermopiles. The first (upper left) was designed for measuring the thermal powers of Sr^{90} samples of 5 to 50 watts. The shaft ΔT is ~1.5°C per watt.

The reproducibility of results obtained with this calorimeter is illustrated by the calibration data given in Table VI-1. No individual value of the calibration constant differs from the average by more than 0.13%. If deviations from the average are treated as random errors,

the 95% confidence limit of a single determination is 0.2%.

The performance of the calorimeter is only moderately sensitive to the rate of flow or to the temperature of the cooling water provided both are constant during any given determination. Variation of the flow rate from 1.1 to 8 liters/min produced a change of only 0.4% in the calibration constant. A change in the cooling-water temperature from 10 to 27°C lowered the constant by 1.4%. A lowering of 1.56% was calculated from the known change of the iron-constantan thermopile and the copper shaft with temperature. The agreement is within experimental error.

The second calorimeter (upper right) was designed for use with Cs^{137} samples of 5 to 50 watts. The cup is constructed of metallic uranium and has walls $1\frac{1}{16}$ in. thick. The gamma-ray absorption is nearly complete; more than 99.8% of the total energy is absorbed.

Because of the heat capacity of the thick-walled cup, a much longer time (7 to 8 hr) is required for this calorimeter to reach steady state than is required for calorimeters designed for use with only alpha- or beta-emitting materials. The results are no less accurate, however, being reproducible within 0.2%.

The third calorimeter (lower left) is designed to allow more rapid measurements of gamma-emitting sources. The thinner walls absorb a known fraction of the radiation (94.85% of the total energy from Cs^{137}). Thus the thermal powers of gamma-emitting samples can be determined in approximately the same time (1.5 to 2 hr) as that required with calorimeters designed for use with alpha- and beta-emitting materials. Some uncertainty is added because of differing degrees of self-absorption of gamma radiation by the samples themselves.

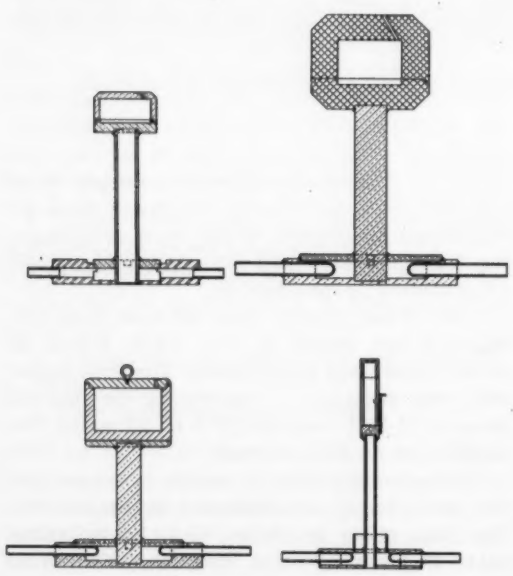

Fig. VI-5 Thermal-conductivity calorimeters.

Table VI-1 CALIBRATION DATA OBTAINED WITH CALORIMETER SHOWN IN UPPER LEFT-HAND CORNER OF FIG. VI-5

Thermal power, watts	Thermopile output, mv	Calibration constant, watts/mv
24.18	6.594	3.667
24.32	6.626	3.671
18.38	5.005	3.673
18.08	4.929	3.669
12.27	3.339	3.674
12.32	3.354	3.675
6.17	1.682	3.676
		3.672 (av.)

The last calorimeter was designed for use with small samples of alpha- or beta-emitting materials held in a standard capsule used for irradiation of samples in the hydraulic tubes of the Oak Ridge Research Reactor. Measurements are accurate to 0.005 watt in the range 1 to 10 watts. A similar calorimeter with thick uranium walls has been constructed for gamma-ray absorption. It has been used primarily for the assay of small Ir^{192} samples.

A calorimeter for assaying alpha-emitting samples of 30 to 300 watts is being made. It will extend the range of sample power that can be measured. Since the shaft has a much larger thermal conductivity than that of any of the calorimeters previously constructed, it will be a test of the degree to which the time required to reach steady state can be reduced by this means.

Flow Calorimeters

Assay of sources in the form of long, thin rods is sometimes necessary. The calorimeters described above are not suitable for such assays, and two calorimeters have been designed, one for alpha- or beta-emitting sources and one for gamma-emitting sources, which

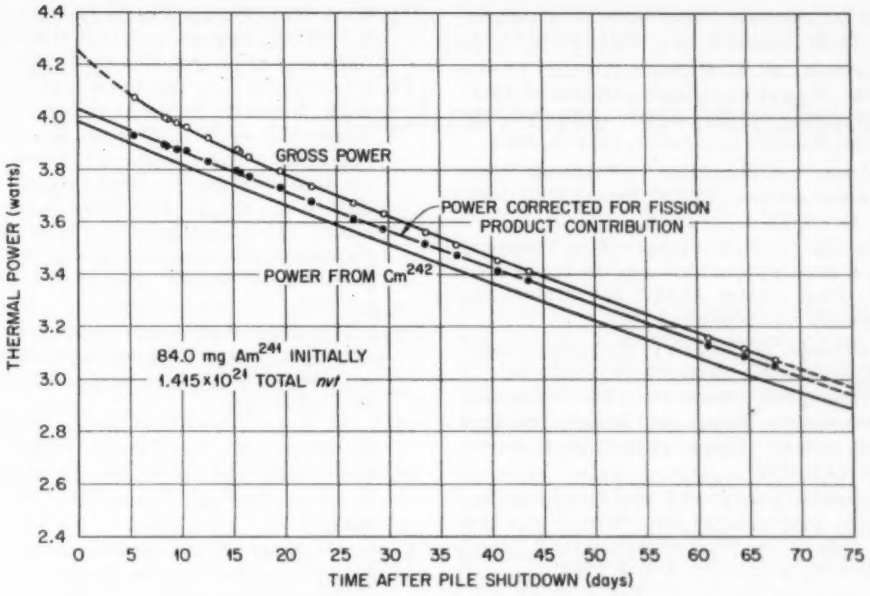

Fig. VI-6 Calorimetric decay study of irradiated Am^{241} (Cm^{242}).

may be used with long sources. The increase in temperature of a constant and accurately measured stream of water is used as a measure of the rate of heat production by the source in these new devices.

Applications of Calorimetry

Calorimetric methods are used for the final check of the magnitude of the heat output of the radioactive heat sources produced by the Isotopes Development Center and are for control analysis during manufacture of the materials of these sources.[17] Calorimetry also provides a convenient measurement of the degree of conversion of samples exposed in atomic reactors. The samples are removed from the reactor, and the thermal power is measured without removing the sample from its container. This method has been used in the curium development program.

One notable project carried out in support of the above work was a decay study of an irradiated Am^{241} sample. Although the thermal power of this sample was largely produced by the Cm^{242} product, significant fractions were produced by fission products and by long-lived transuranic impurities. Ordinarily the quantities of these other radioactive materials present in samples are calculated, so it was desired to check the accuracy of the methods of calculation.

A decay study was carried out over a period of 65 days (Fig. VI-6). The total or measured powers were corrected by the normal method for the contribution of fission-product decay. Since the decline of the corrected power with time could be attributed within experimental error entirely to the decay of Cm^{242} to Pu^{238}, these values could be correlated with time by a least-mean-squares equation, using literature values for the half-lives and energies of decay of Cm^{242} and Pu^{238}. Agreement of this derived equation with the data was excellent; the standard deviation of the points from the equation was only 0.0023 watt, or 0.06% of the average power of the sample.

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Section

VII

Isotopes and Radiation Technology

Applications in Medicine

The literature on medical uses of radioisotopes is briefly reviewed here.* Since developments in this rapidly expanding field have been summarized in various well-documented publications,[1] earlier original literature was not extensively consulted. Only a few of the many published papers are listed here, and emphasis is on recent rather than older work. Since many reports are preliminary and opinions on the effectiveness of some of the isotopes differ widely, future issues of this Review will present various viewpoints on the medical use of radioisotopes, and some of the less well-known isotopes will be reviewed. Readers with publications in this field are invited to send in reprints to ensure coverage.

The medical use of artificial radioactive isotopes began in the late thirties.[2] The increasing interest in them is indicated by the numerous conferences,[3] journals,[4] bibliographies,[5] and reviews[6] partly or entirely devoted to this field. Radioisotopes are used in both diagnosis and therapy (Table VII-1);[7] in the former, in extremely small amounts; in the latter, chiefly in malignant or malignancy-related conditions. Safety in their use is ensured (1) by requiring the licensing of physicians who use them, and then only after special training and (2) by standardization of handling techniques and adoption of special safety regulations by hospitals.[8] Early fears of long-term adverse effects on patients have been tempered by the results of experience. According to Lawrence,[9] for example, cancer has not once been reported as a result of radioactive iodine therapy of thousands of hyperthyroid patients; and in 25 years the survival rate of more than 500 polycythemia vera patients treated with P^{32} has been about the same as that of patients with diabetes or pernicious anemia treated by standard methods. There is wide usage of radioactive iodine for toxic goiter in patients over 45, or P^{32} for polycythemia vera, and of various isotopes for pituitary or thyroid ablations to relieve other conditions.[1]

Among the radioisotopes now used in internal medicine in the largest amounts are I^{131}, P^{32}, Au^{198}, Cr^{51}, Fe^{59}, and Hg^{203}, but many others are used in smaller amounts; large quantities of Co^{60} and Cs^{137} are used in teletherapy units. Many radioisotopes are being studied for potential use, and useful ones are being introduced into the field as new production methods are developed and costs are lowered. Criteria for suitable isotopes include high specific activity, suitable half-life, physiological compatibility with body processes, and the ability to at least partially localize in the tissue being examined or treated. Radioiodine, for example, although not entirely thyroid-specific, is an obvious choice for thyroid conditions, as is calcium for bone. Specificity may sometimes be obtained by incorporation in compounds (e.g., radiocobalt in vitamin B_{12}, which localizes in the liver). An isotope may be localized physically as a wire, as a colloid, or as the recently introduced ceramic beads.[10] Both the physiological and biological half-life must be considered, and selection may be made on the basis of the need for particular radiation characteristics—from a soft-beta all the way to a hard-gamma radiation. In earlier work isotopes with a short physical half-life were found inconvenient to use,[11] but they are finding application now with development of new instrumentation and sources of supply.

* The editors of this Review gratefully acknowledge the many helpful suggestions made by W. G. Myers, M.D., and M. Brucer, M.D., who reviewed a rough draft of the manuscript. Some of their suggestions that could not be readily incorporated will be used in future issues.

Table VII-1 RADIOISOTOPES USED IN DIAGNOSIS AND THERAPY*

Diagnosis

Test	Measurement	Usefulness
I^{131} studies of thyroid function; NaI, usually oral	Percentage of dose concentrated in thyroid; percentage of dose excreted in urine; free and protein-bound I^{131} in plasma	Valuable index of thyroid function; drugs cause interference
I^{131} for diagnosis of thyroid carcinoma; oral	Scanning and external procedures	Relatively little value while normal thyroid gland is present; highly useful in locating well-differentiated metastatic lesions after thyroidectomy
Fe^{59} iron metabolism; usually intravenous iron citrate; oral doses used to study absorption	Serial plasma assays for several hours; red-cell assays for about 10 days; external counts over spleen, liver, and sacrum; blood and fecal assays after oral dose	Helps explain mechanism of some anemias; importance of impaired erythropoiesis vs. hemolysis; oral test indicates absorption defects
Cr^{51} red-cell survival; red cells labeled with Cr^{51} in vitro and injected intravenously	Serial assays of blood (red cells) for two or three weeks; external counts over spleen, liver, and heart; fecal assays for gastrointestinal bleeding	Indicates rate of hemolysis and, to some extent, the role of the spleen in hemolysis
Co^{60}–vitamin B_{12}; oral	Fecal and urinary excretion; sometimes external counts over liver	Shows fundamental defects of pernicious anemia, even if patient is not in hematologic relapse
I^{131}-labeled rose bengal; intravenous	Rates of removal from bloodstream and concentration in liver	An index of liver function and biliary patency
I^{131}-labeled fat; oral	Blood, urinary, and fecal levels	Information on intestinal absorption and pancreatic function
Cr^{51}, P^{32} red-cell mass; I^{131} albumin-plasma volume; Na^{24}, K^{42} for sodium and potassium spaces	All based on dilution principle; solution of isotope (or suspension of red cells tagged in vitro) given intravenously, and, after mixing in the body, blood concentration is determined	These tests give data on red-cell and plasma volumes, fluid balance, and electrolyte status; may be helpful in presurgical evaluation and study of metabolic and endocrine disorders

Therapy

Treatment	Patients treated	Usefulness
I^{131} as NaI; usually oral	Uncomplicated Graves' disease in patients over 40; complicated Graves' disease, any age	General results excellent; multiple doses needed in some cases; significant incidence of early or delayed myxedema
	Toxic nodular goiter in selected patients	Eventual control of toxicity in most cases; may require repeated doses; concern over malignancy already present at onset, plus desire for rapid result, may recommend surgery for patients who can be well prepared
	Thyroid carcinoma in selected patients	For treatment of relatively small group of patients with non-resectable lesions showing ability to concentrate the isotope
	Certain patients with angina pectoris or intractable congestive heart failure	Clinical improvement in significant percentage

(Table continues.)

Table VII-1 (Continued)

P^{32} as soluble phosphate; oral or intravenous	Chronic granulocytic leukemia	Generally good results
	Chronic lymphocytic leukemia	Useful, often in conjunction with local irradiation
	Polycythemia vera	Good results in patients not suitable for phlebotomy alone
	Lymphosarcoma; multiple myeloma; diffuse bone metastases	Occasionally some value
	Acute leukemia; Hodgkin's disease	Rarely useful
Colloidal Au^{198}, P^{32}, and Y^{90}	Intracavitary in patients with effusions and ascites caused by malignant tumors	Symptomatic value; partially supplanted by nitrogen mustards
	Intravenous for granulocytic leukemia and diffuse lymphoma of liver	Very limited value compared with other forms of therapy
	Direct injection into tumor in certain cases of prostate carcinoma; attempt to radiate lymphatic channels	Believed useful if properly used in carefully selected cases
Co^{60}, Cs^{137}, Ra^{226}, Rn^{222}, and Sr^{90} for implantation and brachytherapy	Certain localized tumors, especially carcinoma of cervix, head, and neck	Useful in skilled hands for properly selected patients
Co^{60}, Cs^{137} for teletherapy	For all lesions suitable for external radiation therapy	Highly useful and practical

* Reprinted with minor style changes with the kind permission of publishers, from G. A. Andrews, Medical Students and Radioisotopes, *The New Physician*, 9(12): 23-27 (1960).

Iodine

I^{131} (8.1 Days, 0.33- and 0.61-Mev β's, 0.36- and 0.64-Mev γ's). Iodine-131 is most commonly used for diagnosis of thyroid disorders.[1] Other uses include incorporation in human serum albumin for determination of the total circulating plasma volume, placentography,[12] estimation of mitral valve size,[13] and detection of right-to-left cardiac shunts. There has been partial success in localizing brain tumors with I^{131}-labeled compounds, and I^{131}-labeled methyl iodide has been used for detecting left-to-right cardiac shunts.[14] Rose Bengal containing I^{131} is useful for liver-function tests and for blood-flow tests[15] and has been suggested for detecting liver metastases that are not obvious from surgery.[16] Recent reports indicate the Rose Bengal–I^{131} hepatoscan to be a satisfactory technique.[17] Other I^{131}-labeled compounds reported useful are Hippuran for renal tests[18] and for determining cerebral circulation time;[19] Diodrast for evaluating venous diseases of the lower extremities;[20] sulfobromophthalein for liver-function tests;[21] and compounds for studying fat metabolism.[22] An I^{131}-labeled compound demonstrated 200-ml pericardial effusions in a simulated system,[23] and an accurate and reproducible method for insulin assay with I^{131} has been reported.[24]

In therapy, I^{131} has been used for nontoxic[25] as well as toxic goiter.[1] It is used as a palliative in thyroid carcinoma, and an I^{131}-tagged fat-soluble medium, which accumulates in regional lymph nodes, is potentially useful for irradiation of neoplasms.[26] Thyroid ablation by I^{131} in more than 1000 patients improved angina pectoris in 75% and congestive heart failure in 60% of those afflicted.[27] Fortunately, gamma irradiation of the ovaries from urinary excretion of administered I^{131} is considered negligible.[28]

I^{125} (57.4 Days, No β, 0.035-Mev γ). Iodine-125, which is easily produced by neutron bombardment of Xe^{124}, is becoming increasingly valuable in diagnosis. Irradiation of a patient by I^{125} is about one-half[29] that from I^{131}, assuming that the biological half-time is equal to the physical half-life. Good thyroid and liver scans have been made with it,[30] and Hippuran labeled with I^{125} has been used for renograms.[31] As a tag for serum albumin and oleic acid and in the salt NaI^{125}, I^{125} has given good results in experimental blood volume, absorption, and thyroid tests,[32] and it has been suggested for indicating the extent of arteriosclerosis.[33] In a promising new technique for lipid digestion and absorption studies, a compound labeled with I^{131} and another labeled with I^{125} are administered simultaneously.[34] The promising use of I^{125} in

radiography[35] is indicated by a photograph of the bones of a hand made with this isotope (Figs. VII-1 and VII-2). Exposure to both patient and technician is low because of the ease of shielding against the 27.2- and 35.4-kev photons. Portable X-ray equipment is being developed with I^{125} sources.[36]

I^{132} (2.3 Hr, 1.5- to 2.1-Mev β's, 0.53- to 0.95-Mev γ's). The use of I^{132}, which is easily recovered from a tellurium source, would decrease patient exposure to 1 to 2% of that from I^{131} and would allow repeated administration.[37] Thyroid function after I^{131} therapy may be determined[38] by observing the absorption of I^{132}, whose higher energy gammas are easily detected in the presence of residual I^{131}.

I^{123} (13.3 Hr, No β, 0.16-Mev γ). With I^{123}, exposure of a patient would be only 1 to 5% of

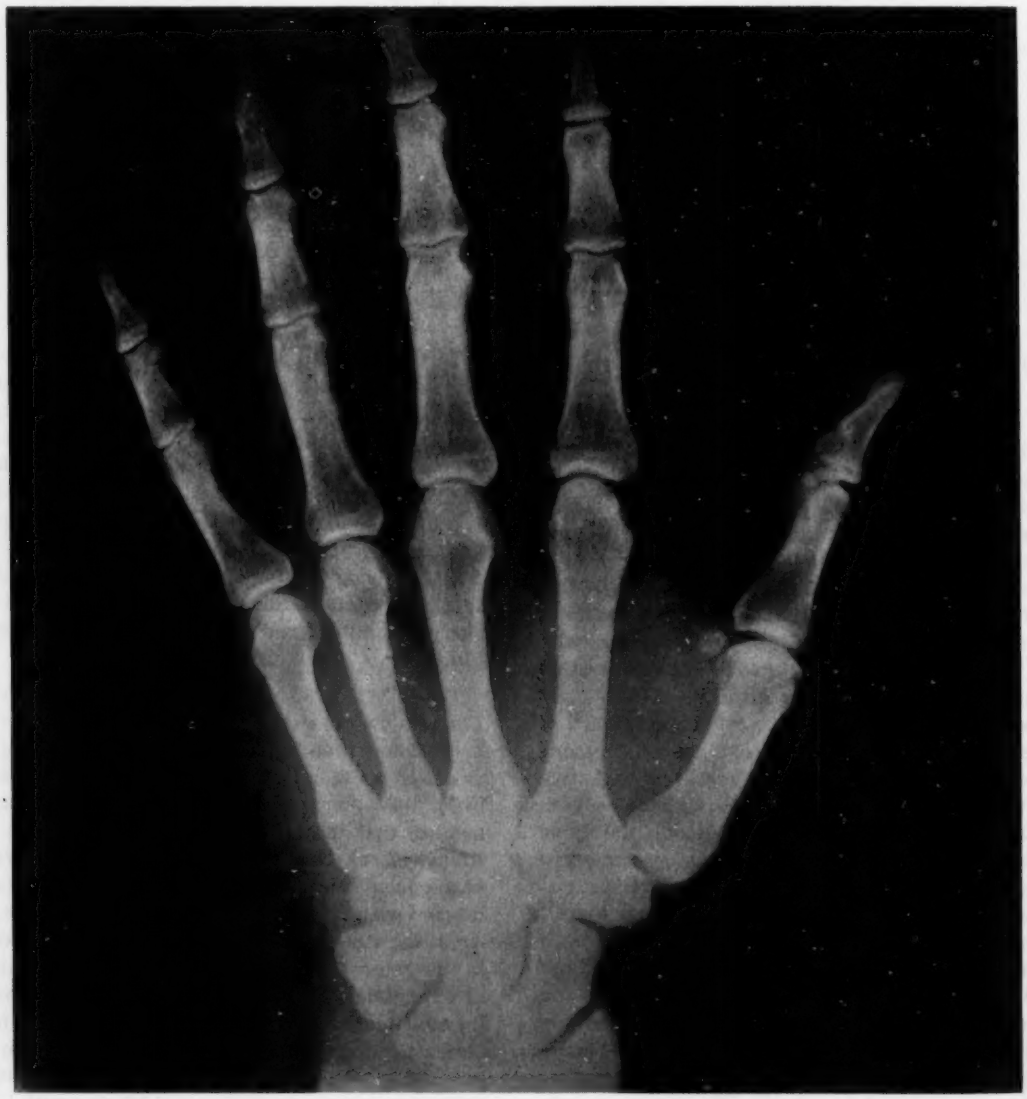

Fig. VII-1 Radiograph of a hand made with a 1.96-curie I^{125} source at 12 in. Intensifying screen used. Time: 2 min. Dose: 9.5 mr. Film: Kodak Royal Blue X Ray.

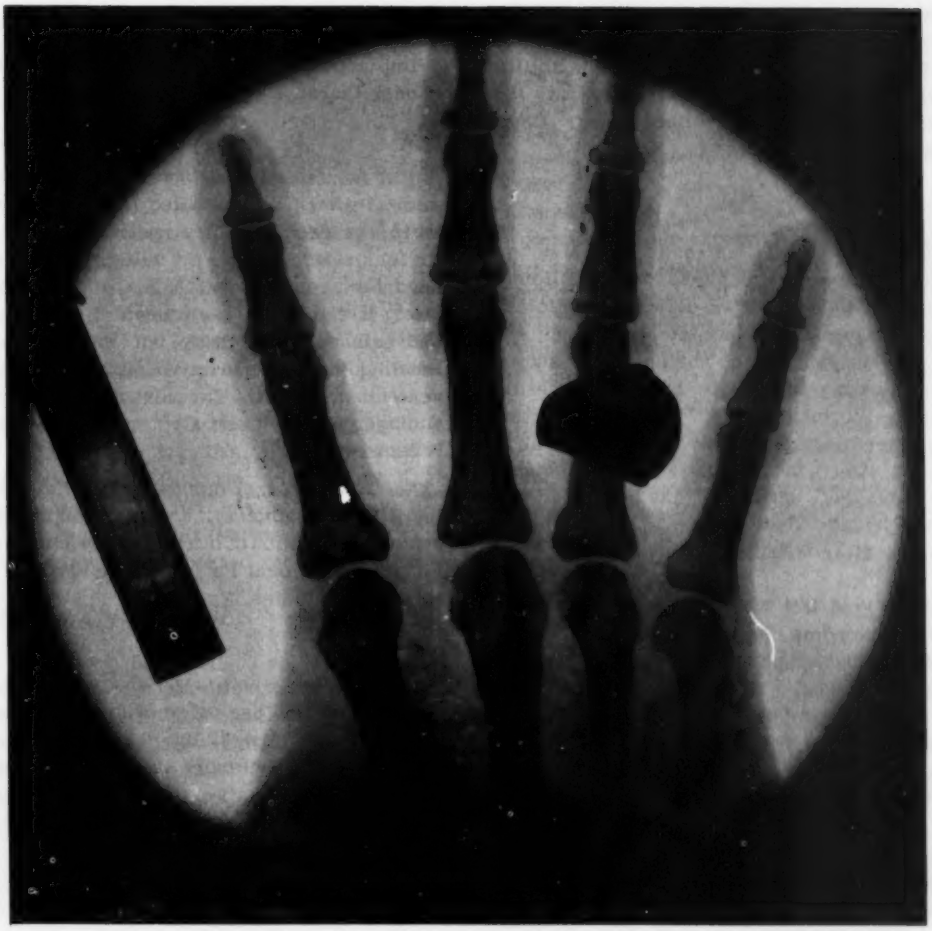

Fig. VII-2 Radiograph of a hand and a pocket dosimeter made with a 1.96-curie I^{125} source at 6 in. No intensifying screen. Time: 30 sec. Dose: ~9 mr. Film: Polaroid type 3000X.

that from I^{131}. Experiments on preparation of curie amounts are projected.[39]

I^{124} (4 Days, 1.53- and 2.13-Mev β's, 0.6- and 1.72-Mev γ's). Iodine-124 has been suggested for greater uniformity of dose in thyroid carcinoma.[40]

Phosphorus

P^{32} (14.3 Days, 1.71-Mev β, No γ). The largest use of P^{32} is in treatment of polycythemia vera and chronic leukemia.[1] It has not been very useful in acute leukemia. Blotting paper soaked with radioactive sodium phosphate was used in early applications for skin lesions, and activated red phosphorus incorporated in polyethylene was used in later work. Metastatic bone cancer has been treated with P^{32} as the phosphate.[41]

Colloidal chromic phosphate-P^{32} and Au^{198}, as well as nitrogen mustard, for control of malignant intracavitary effusions are compared in a recent review[42] of 222 cases treated with P^{32} and 2110 with Au^{198}. Colloidal chromic phosphate-P^{32} has been used for palliation of cancer, and colloidal zirconyl phosphate-P^{32} administered intravenously to more than 250 patients with chronic leukemia and lymphomas is reported to have resulted in prolonged remissions and long treatment-free periods.[43]

In diagnostic applications, an in vivo test for human spermatogenesis using sodium phosphate-P^{32} has been described,[44] and the ability of newly proliferating cells to concentrate P^{32} has been used to differentiate benign from malignant ocular lesions.[45]

Gold

Au^{198} (2.7 Days, 0.97-Mev β, 0.41-Mev γ). Gold-198 is most commonly used as a colloid,[1] which has 0.003- to 0.007-μ-diameter particles. When this material is administered to a patient and a scan is performed, normal liver shows an even gradation of activity, but lesions result in activity concentrations.[46] Diagnostic uses include determination of liver blood flow,[15] and, in regions where the parasitic taenia *Echinococcus granulosus* occurs, it has been used for localizing hydatid liver cysts and for following up healing after drug administration or surgery.[47]

Colloidal gold has been used in treating both primary carcinoma and metastases.[48] Transportation to the lymph nodes is hastened if the particles are coated with silver,[49] but radioactive colloidal gold has been injected directly into the lymphatics.[50] Other uses are in treating leukemia[51] and in controlling malignancy-induced intracavitary fluids.[52]

Interstitial implants of Au^{198} seeds have been used for hypophysectomy and for palliation of metastatic carcinoma.[53] The hypophysis implantation requires about 30 min. Problems in surgical and radiotherapeutic pituitary ablation are discussed in recent reviews.[54]

Chromium

Cr^{51} (28 Days, No β, 0.32-Mev γ). Hexapositive Cr^{51}, in $Na_2Cr^{51}O_4$, attaches to the red cells and may be used to determine such quantities as red-cell survival time.[1] A new instrument[55] for accurate blood-volume measurement uses Cr^{51}-tagged red cells or I^{131}-albumin. Such cells have been suggested for determination of cardiac output and for placental localization (no activity is transferred to the fetal circulation) and are the basis for a simple method for quantitation of menstrual blood loss[56] and for spleen studies.[57] Tripositive chromium (as $Cr^{51}Cl_3$) in solution attaches to plasma proteins and is useful for determining plasma volume. Serum albumin labeled with Cr^{51} is easily prepared and has been introduced for demonstrating gastrointestinal protein loss.[58]

Implants of Cr^{51} have been recommended for cancer therapy.[59]

Iron

Fe^{59} (45 Days, 0.26- and 0.46-Mev β's), 1.1- and 1.3-Mev γ's). Detailed methods for Fe^{59} tests are described in a recent brochure.[60] As the chloride, citrate, or sulfate, Fe^{59} has been used for iron-absorption and -utilization studies.[61] It has also been used for determining red-cell survival time, but the necessity of labeling the cells in vitro makes it less convenient than Cr^{51}. The advantages and disadvantages of Fe^{59} and Cr^{51} are discussed in a recent review.[62]

Fe^{55} (2.94 Years, 0.0059-Mev X Ray Following Electron Capture). Comparison of the percentage utilization of intravenously administered Fe^{59} to that of oral Fe^{55} has been used for iron-absorption studies.[63]

Mercury

Hg^{203} (47 Days, 0.21-Mev β, 0.28-Mev γ). In addition to the use of mercury isotopes[64] in tracing kidney drugs, Hg^{203} has been used for localizing brain tumors and for determining cerebral circulation time.[65] Neohydrin and Chlormerodrin labeled with Hg^{203} have been used for kidney examinations.[66]

Boron

B^{10}. When the stable isotope B^{10} is deposited in brain-tumor tissue and activated by neutron bombardment, the released alpha radiation is effective for control and palliation.[67] By this treatment[68] cells of transplantable sarcomas in mice have been killed, and cancer has disappeared in 2 to 4 weeks. This method of treatment is not new, but basic techniques are still being investigated.[69]

Fluorine

F^{18} (1.87 Hr, 0.65-Mev Positron Emission). Fluorine-18 has been used as a replacement for Ca^{47} and Sr^{85} in bone scanning.[70] It has also found application in thyroid studies[71] and in locating brain tumors.[72]

Selenium

Se^{75} (120 Days, No β, 0.14- to 0.28-Mev γ's). The high specificity of Se^{75}-selenomethionine

for the pancreas suggests that this compound might be useful in early detection of pancreas carcinoma.[73]

Krypton

Kr^{85} (10.6 Years, 0.67-Mev β). Because of its low solubility, injected or inhaled Kr^{85} is useful in determining cerebral and coronary blood flow and cardiac and pulmonary shunts.[74] For example, if a right-to-left cardiac shunt is present, activity in the arterial system rises rapidly when Kr^{85} is given intravenously.[75] If a left-to-right shunt is present, the Kr^{85} concentration in the pulmonary artery reaches a value 20 times that in a systemic artery following inhalation of this gas.[76] A simple technique for studying pulmonary function with Kr^{85} has been devised,[77] and a new technique for rapid and accurate measurements of serial heart blood flow was reported recently.[78]

Strontium and Yttrium

Sr^{90} (28 Years, 0.54-Mev β, 2.27-Mev β from Y^{90} Daughter, No γ). In an external source containing Sr^{90} designed for use in ophthalmology, the Sr^{90} beta particles are filtered out so that only the Y^{90} betas actually reach the lesion. The lens is not irradiated.[1]

"Radiation curettage," destruction of the uterine mucosa by radiation from a Sr^{90} source placed in the uterine cavity, has been suggested for control of endometrial bleeding when no malignancy is indicated.[79]

Sr^{85} (65 Days, No β, 0.5-Mev γ). Strontium-85 has been used for localizing bone tumors[80] and for studying bone metabolism. Because of its shorter half-life, irradiation of the body with Sr^{85} is less than with Sr^{90}. Its gamma radiation can be detected externally.

Sr^{87m} (2.8 Hr, 0.39-Mev γ). This isotope, which can be obtained quickly, as needed, from 80-hr Y^{87}, has been mentioned[81] for possible use in obtaining information available in a short time; for example, circulation time, cardiac output, and various clearances, and in radiography.

Y^{90} (64.5 Hr, 2.27-Mev β). Injected ceramic Y^{90} particles approximately 60 μ in diameter, which are trapped in capillaries of the lung, liver, and other organs, have been used for cancer therapy.[82] Thirty-five patients with Parkinson's disease[83] were improved by brain implants of ceramic Y^{90} beads. Carrier-free Y^{90} may be eluted[84] with citrate from a cation-exchange-resin preparation of Sr^{90}. It may be infiltrated as a solution, implanted in a plastic filament, or applied in a bag or absorbed in blotting paper.[85]

Because it is a pure beta emitter, colloidal Y^{90} results in less exposure of hospital personnel than Au^{198}, and it has been used for control of malignancy-induced effusions.[86] Complete pituitary destruction has been reported as a result of packing the fossa, following surgery, with powdered Y^{90} dispersed in wax.[87] For superficial radiation therapy, Y^{90} has been incorporated in small plates, which can be selected to cover an irregular area.[88] Transperineal injection of Y^{90} has been used for prostatic cancer.[89] Intravenous Y^{90} gave good results in eight polycythemia vera patients.[90]

Calcium

Ca^{45} (164 Days, 0.25-Mev β, No γ). Calcium-45 has been used for studying calcium metabolism, Paget's disease of bone, and hypoparathyroidism.[91]

Ca^{47} (4.7 Days, 0.66- and 1.94-Mev β's, 0.48- to 1.3-Mev γ's; Accompanied by 3.5-Day Sc^{47} Daughter with 0.46- and 0.62-Mev β's and 0.175-Mev γ). Possible medical uses of the now-available Ca^{47}, which is of interest because of its high gamma energy, are being extensively investigated.[92]

Cobalt

Co^{60} (5.3 Years, 0.31-Mev β, 1.17- and 1.33-Mev γ's). By far the largest use of Co^{60} is in teletherapy units,[1b,1c,93] descriptions of which may be obtained from various equipment manufacturers. Such units[94] were widely distributed by 1959 (Table VII-2). In 1962 they were reported to number 1400, of which 550 are in the United States and Latin America.[95] They are reported[96] to be in use in Communist China and have been referred to as the most important product of Atomic Energy of Canada Limited.[97] A nonroutine use of Co^{60} teletherapy is a possible new treatment for leukemia in which blood, circulated outside the body through a tube, is exposed to Co^{60} radiation.[98]

Cobalt-60 needles have been implanted in tumors, and seeds enclosed in nylon tubing have been used in interstitial therapy and in

Table VII-2 RADIOISOTOPE TELETHERAPY SOURCE DISTRIBUTION*

Location	No. of Units Ir^{192}	Cs^{137}	Co^{60}
Africa			
Algeria			1
Union of South Africa			1
America			
Argentina			3
Brazil			8
Canada		2	24
Chile			2
Cuba			2
El Salvador			1
Mexico			12
Peru			1
Puerto Rico			2
United States		6†	264†
Uruguay			2
Venezuela			5
Asia			
Burma			1
Ceylon			1
China (Taiwan)			1
Hong Kong			1
India			3
Indonesia			1
Iran			1
Israel			3
Japan			200‡
Lebanon			1
Philippines			4
Thailand			1
Europe			
Austria			2
Belgium			2
Denmark			2
Finland			1
France		1	40
Germany (Fed. Republic)			4
Greece			3
Hungary			1
Italy			26
Monaco			1
Netherlands			4
Norway		1	
Portugal			1
Spain			5
Sweden			7
Switzerland			4
United Kingdom	1	8	33
Yugoslavia			1
Oceania			
Australia			4
New Zealand			2
Total§	1	18	689

* Reprinted, with minor style changes, from Ref. 94.

† These figures represent the number of licenses issued as of Aug. 1, 1959; they do not include such teletherapy installations as those at Oak Ridge Institute of Nuclear Studies and Argonne Cancer Research Hospital, for which no license was required.

‡ Estimated, mainly low-curie units.

§ Including only countries for which information is available.

wounds after surgery for a malignant condition.[99] Spheres of Co^{60}, gold plated to prevent dusting, may be placed in hollow organs such as the uterus or bladder.[100] Cobalt-60 may be incorporated in plastic and molded to fit irregular areas such as the oral cavity.[101]

Vitamin B_{12} labeled with Co^{60} has been used in diagnosing pernicious anemia[102] and in localizing liver tumors.[103]

Other Cobalt Isotopes: Co^{56} (77.3 Days, 0.44- and 1.46-Mev β's, 0.85- to 1.75-Mev γ's); Co^{57} (270 Days, No β, 0.014- and 0.12-Mev γ's); Co^{58} (71 Days, 0.48-Mev β, 0.81-Mev γ). Various other cobalt isotopes have been used[1] to label vitamin B_{12}. Cobalt-57 is reported[104] to give the lowest irradiation of a patient. Labeled vitamin B_{12} (Co^{57}) has been shown to concentrate in the parathyroid in dogs[105] and is therefore potentially useful in parathyroid conditions.

Cesium

Cs^{137} (30 Years, 0.51- and 1.18-Mev β's, 0.662-Mev γ from Daughter Ba^{137}). This isotope, which is recovered as a by-product from the processing of nuclear reactor fuel, is a relatively new isotope, but many Cs^{137} teletherapy units are in use (Table VII-2). The long half-life and easier shielding of the gamma rays are the principal advantages over Co^{60}. Typical units have been described elsewhere.[1c,93a,106]

Iridium

Ir^{192} (74 Days, 0.67-Mev β, 0.30- to 0.47-Mev γ's). Radioiridium teletherapy is an old idea.[107] A new 100-curie Ir^{192} irradiation unit weighs only 38 lb; a 10-curie source weighing 28 lb is also available.[108] A unit with interchangeable ends of different sizes and shapes, for interorificial therapy, has been described recently.[109]

The half-life of Ir^{192} is about 5% that of Co^{60}, and its less-penetrating gamma rays allow greater local absorption and necessitate less protection for hospital personnel. Internally, it has been used as seeds embedded in nylon ribbon or as needles.[110] Experimentally, an Ir^{192}-labeled resin suspension has been used with external counting for determining gastric emptying rates.[111]

Arsenic and Copper

As^{74} and Cu^{64} (17.4 Days and 12.8 Hr, Respectively; Positron Emitters). These isotopes have been used for localizing brain tumors.[112]

As^{76} (26.5 Hr) and As^{72} (26 Hr). Arsenic-76 has been used for brain-tumor localization, as has As^{72}, which yields nearly three times as many positrons per average disintegration[113] as As^{74}.

Gallium

Ga^{72} (14.3 Hr, 0.64- and 0.96-Mev β's, 0.63- to 2.20-Mev γ's). Gallium-72 has been used to localize malignant bone tumors by external measurements, but the short half-life results in a concentration of this chemically toxic material which may perhaps be higher than is warranted by the radiation delivered.[114]

Ga^{68} (68 Min, 1.9-Mev β, 1.1-Mev γ). Gallium-68 has been suggested for such applications as brain-tumor localization by annihiscopy. A source from which Ga^{68} can be easily and rapidly obtained has been designed.[115]

Palladium

Pd^{109} (13.5 Hr, 1.03-Mev β) and Pd^{103} (17 Days). A mixture of these two isotopes has been used for interstitial irradiation.[116]

Tantalum

Ta^{182} (115 Days, 0.36- to 0.51-Mev β's, 1.12- and 1.22-Mev γ's). Tantalum-182 wires have been used as tissue implants in control of malignant conditions.[117]

Ruthenium-Rhodium

Intragastric beta irradiation with Ru^{106}-Rh^{106} has been tried.[118]

Lutetium

Lu^{177} (175 Days; Also 6.8 Days, 0.50-Mev β, 0.18- and 0.38-Mev γ's). Lutetium-177 has been suggested for infiltration to lymph nodes involved in malignant disease.[119]

Sulfur

S^{35} (87 Days, 0.167-Mev β, No γ). Sulfur-35 has been used for determining extracellular water and as a therapeutic agent in chondrosarcoma and mycosis fungoides.[120] Labeling of Congo red dye with S^{35} decreases the amount of dye needed for diagnosis of amyloidosis, eliminating the side effects that occur in some patients.[121] Methionine labeled with S^{35} has been suggested for use in detecting pancreas tumors.[122]

Sodium

Na^{22} (2.6 Years, 0.54-Mev β, 1.3-Mev γ) and Na^{24} (15 Hr, 1.4-Mev β, 2.7- and 1.4-Mev γ's). These isotopes have been used for measuring extracellular water, sodium space, and exchangeable sodium; for determining when skin grafts are complete; and for cancer therapy.[1,123] Because of its shorter half-life, Na^{24} has advantages for some uses.

Potassium

K^{42} (12.4 Hr, 3.6-Mev β, 1.5-Mev γ) and K^{43} (22 Hr, 0.3- and 0.27-Mev β's, 0.37- to 0.62-Mev γ's). These isotopes have been used to localize brain and breast tumors, measure exchangeable potassium, and study potassium uptake by red blood cells.[1,124] Less shielding is required with K^{43} because of its lower energy gamma radiation.

Rubidium

Rb^{86} (19 Days, 1.78- and 0.72-Mev β's, 1.1-Mev γ). Since rubidium acts similarly to potassium and has a more convenient half-life than K^{42}, it has been suggested for use in determining exchangeable potassium.[124c] However, it must be remembered that Rb^{86} is *not* a potassium isotope. Myocardial scanning tests have been made with Rb^{86} in dogs.[125]

Bromine and Chlorine

Br^{82} (35.7 Hr, 0.44-Mev β, 0.55- to 0.78-Mev γ's). Bromine-82 ratios in blood and spinal fluid have been used to differentiate between tuberculous and nontuberculous meningitis.[126] For determining extracellular fluid,[127] it has advantages over the 37-min Cl^{38} or the 3×10^5-year Cl^{36} (Ref. 1*a*, p. 233).

Solutions of Br^{82}, enclosed in a rubber balloon, were used in early work[1,123e] for treating carcinoma of the bladder.

Silver

Ag^{111} (7.5 Days, 1.05- and 0.69-Mev β's, 0.34-Mev γ). Colloidal Ag^{111} was one of the early radioactive colloids used.[128] Silver-111, injected into the bloodstream as a protein-silver complex, is removed less rapidly by a cirrhotic liver than by a normal one.[129]

Zinc

Zn^{63} (38 Min, 1.38- to 2.34-Mev β's, 0.67- and 0.96-Mev γ's). Zinc-63 was used in early work for control of malignant effusions.[1]

Zn^{65}. The use of Zn^{65} in studying zinc metabolism was reported recently.[130]

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Section

VIII

Isotopes and Radiation Technology

Safety

The U. S. Atomic Energy Commission (AEC) is seriously interested in the safety aspects of the isotopes program. Their contracts include studies of both shipping casks and radioactive sources and source capsules as evidence of their desire to reduce radiation hazards. Battelle Memorial Institute, for example, has embarked upon a program to develop safety performance criteria for sealed sources.[1] Information has been accumulated on source applications, designs, fabrication techniques, and testing procedures. Existing regulations, specifications, and performance criteria were also surveyed, compiled, and evaluated. This work and the work at Oak Ridge National Laboratory (ORNL) on actual testing of commercial sources will be reported in more detail at a later date.

Exposure of Personnel Handling Radioisotopes

The results of a survey[2] of persons working with radioisotopes indicated that, in eight categories of use (Table VIII-1), the annual average external radiation exposure is probably not more than 1250 mrem. The attendant risks to personnel are concluded to be negligible compared to risks of everyday life (Table VIII-2).

The survey was conducted by sending out questionnaires to various establishments licensed to use radioisotopes in the manufacture of a commercial product and in medical diagnosis and treatment and to universities. The persons included in the results were those actually working with or handling radioactive elements. Of 253 licensees replying, represent-

Table VIII-1 WHOLE-BODY RADIATION EXPOSURES OF PERSONNEL HANDLING RADIOISOTOPES

Field of isotope use	No. of licensees	Exposure, mrem		No. of persons exposed					% exposed to 0 to 600 mrem
		Range	Average	0 to 600 mrem	601 to 4999 mrem	5 to 11.9 rem	12 to 27.5 rem	Total	
Medical diagnosis and treatment									
X rays or Ra only	12	0 to 3552							
All sources	94	0 to 12,630	461	914	264	6	1	1185	77
Industrial radiography									
Radioisotopes only	17	0 to 2130	253	38	6	0	0	44	86
All sources	47	0 to 7400	472	192	43	2	0	237	81
Thickness, level, or density gaging	19	0 to 4035	604	59	45	0	0	104	57
Universities	24	0 to 3516	178	1259	58	0	0	1317	95
Isotopes* processing and resale	24	0 to 27,453	1726	187	37	21	6	251	75
Industrial tracers	6	0 to 1280	244	89	5	0	0	94	95
Radiation effects studies	10	0 to 2140	112	416	23	0	0	439	95
Static elimination and ionization sources	--	--	--	--	--	--	--	--	--
Total	253			3297	531	29	7	3864	85

*Beta doses were significant in this use category; consequently the tabulated exposures overestimate the whole-body dose.

Tablve VIII-2 COMPARISON OF RISK OF RADIATION EXPOSURE WITH EVERYDAY RISKS OF LIFE

Radiation Exposure

Field of isotope use	1960 exposed population*	No. of persons exposed		Exposure probability	
		5 to 12 rem	12 to 27.5 rem	5 to 12 rem	12 to 27.5 rem
Medical diagnosis and treatment	1378	6	1	4.3×10^{-3}	7.2×10^{-4}
Industrial radiography	281	2	0	7.1×10^{-3}	$<3.6 \times 10^{-3}$
Isotope processing and resale	251	21	6	8.4×10^{-2}	2.4×10^{-2}
All	3864	29	7	7.5×10^{-3}	1.8×10^{-3}

Everyday Risks of Life

Occurrence	Age group, years	1960 or 1961 exposed population	No. of occurrences	Probability of occurrence
Fatal accidents	15 to 64	1.1×10^{8}	5.2×10^{4}	4.9×10^{-4}
Fatal motor vehicle accident	15 to 64	1.1×10^{8}	2.8×10^{3}	2.4×10^{-4} †
Drowning fatalities	15 to 64	1.1×10^{8}	3.9×10^{3}	3.6×10^{-5}
Cancer fatalities	15 to 64	1.1×10^{8}	1.2×10^{5}	1.1×10^{-3}
Home accidents resulting in insurance claims	All	6.0×10^{5}	1.6×10^{4}	2.7×10^{-2}
Sports and recreation accidents resulting in insurance claims	All	6.0×10^{5}	1.0×10^{4}	1.7×10^{-2}
Civilian unemployment	14 or older	7.3×10^{7}	4.0×10^{6}	5.5×10^{-2}

*Age group 18 to 65 years.
†Sic.

ing 3864 persons, 85% received whole-body doses of 600 mrem or less. The exposure data were taken from film-badge dosimeters.

Source Testing

By Karl W. Haff

The stainless-steel capsule of a nine-year-old $Cs^{137}Cl$ source showed no corrosion or deterioration when examined visually and metallurgically.[3] This source, the first $Cs^{137}Cl$ source of greater than 1000 curies built by ORNL, was made in March 1954 and had been used in a teletherapy machine at the Oak Ridge Institute of Nuclear Studies since 1955. It contained two 1.250-in.-diameter CsCl pellets, of 0.531 and 0.504 in. height, 33.07 and 27.86 g weight, and 760 and 780 curies activity, respectively.

The source (Fig. VIII-1) was enclosed in two concentric type 316 stainless-steel capsules, each with a 0.020-in.-thick window. The inner capsule was 2.575 in. long, 1.500 in. in outside diameter, 1.260 in. in inside diameter, and was plugged with a type 316 stainless-steel plug 1.5 in. long and sealed with Easy-Flo silver solder. The outer capsule was 3.265 in. long, 1.750 in. in outside diameter, 1.520 in. in inside diameter, and was plugged with a type 410 stainless-steel threaded plug and sealed with All-State No. 430 silver bearing solder.

When the source was removed from the teletherapy machine head, the outer capsule appeared in excellent condition, with no signs of corrosion or discoloration. A smear of the outer surface showed an activity of 1920 dis/min. No bubbles appeared when the source was immersed in ethylene glycol in a glass vacuum vessel (sensitivity of method 5×10^{-5} cm^3/sec), and the pressure above the source was decreased to −25 in. Hg (about 5 in. Hg absolute).

During sawing off of the outer capsule, a corner of the inner capsule was inadvertently cut off, but no other leaks were observed in the vacuum leak test. A smear showed a high activity, 5×10^{5} dis/min, because of this sawed-

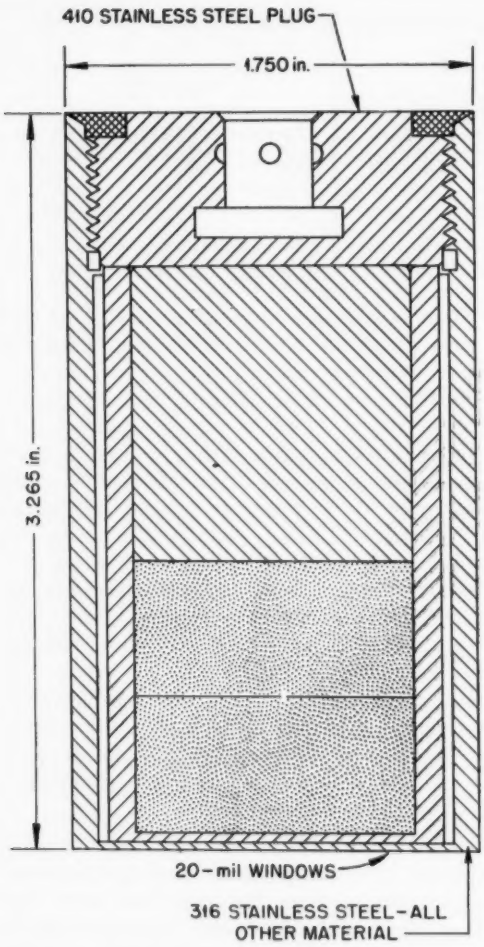


Fig. VIII-1 Oak Ridge Institute of Nuclear Studies 1540-curie $Cs^{137}Cl$ source.

off corner. Visual examination of the exterior surface of the capsule showed no evidence of corrosion or deterioration.

When the inner capsule was opened, the source pellet surface appeared similar to that of a new pellet. It was white to gray in color, with a mottled brown streak near one edge, and the edges of the pellet and the inner wall of the capsule at the edge of the pellet were distinctly brown. However, this discoloration was probably caused by heating during the original sealing of the capsule since, when the pellets were removed, the upper edge of the pellet and the adjacent area on the capsule were seen to be the only affected areas. The inner surface of the capsule and the window area were as bright

Fig. VIII-2 Source capsule before being sectioned for metallurgical studies.

Fig. VIII-3 Transverse section of wall of Oak Ridge Institute of Nuclear Studies source capsule, inside diameter.

and shiny as new material, and inspection under 80× magnification did not show any pitting or cracking.

The capsule was decontaminated by ultrasonic cleaning in water and photographed (Fig. VIII-2). Metallurgical studies of sections at 100× magnification showed no evidence of change in crystalline structure or deterioration of the capsule wall window. Sections are shown in Figs. VIII-3 to VIII-5.

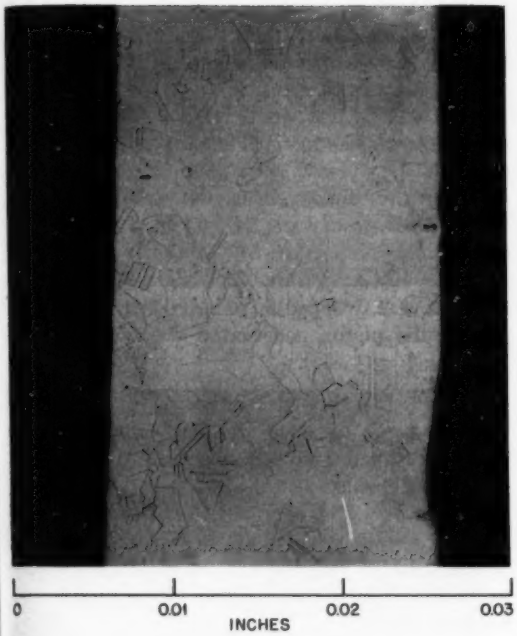


Fig. VIII-4 20-mil window sections (scale on bottom inside of window is to left).

Fig. VIII-5 Longitudinal section of wall of Oak Ridge Institute of Nuclear Studies source capsule, inside diameter.

Shipping-Cask Testing

By Karl W. Haff

The results of experiments performed at ORNL indicate that unprotected lead-filled steel casks weighing up to 6 tons will meet structural requirements of the Code of Federal Regulations, Title 10, Part 72, with the exception of the 6-in.-diameter puncture test. Since the results of a horizontal or piston drop can be estimated with a reasonable degree of accuracy, casks can probably be designed to meet the piston requirements. It is doubtful, however, that such casks would meet fire requirements. Small radioisotope shipping casks that weigh up to 1.5 tons and are approximately 20 in. in diameter can be protected from both impact and fire with low-cost wooden shields.

Lead-filled steel casks and wooden fire and impact shields have been experimentally tested by the Isotopes Development Center at ORNL as part of a continuing program to improve radioactive-material shipping procedures and packaging. The program has included both structural and fire-resistance tests, and experience gained has set the standards for radioisotopes packaging and shipping throughout the United States and the world. Since 1946, the beginning of the radioisotopes program at ORNL, more than 176,000 (as of May 31, 1963) shipments containing approximately 2.2 million curies have been made without a single accident in which a release of radioactivity was involved. However, with the trend in radioactive-material shipments to larger curie content per package, the need of greater safety in package design and shipping has become more apparent. Proposed and existing AEC and Interstate Commerce Commission (ICC) regulations are given in Refs. 4 and 5.

The testing program for fuel shipping casks was discussed in a recent issue of *Nuclear News*.[6]

Lead-Filled-Cask Tests. 1. Structural Tests.[7,8] Forty-one drop tests have been made with five lead-filled steel casks weighing 2725 lb each and two weighing 11,760 lb each. A lead-filled steel cask is shown diagrammatically in Fig. VIII-6. Prior to a drop, each model cask was instrumented with two piezoelectric accelerometers, 12 dynamic strain gages, and compressometers to measure inner wall deflections. Some casks were covered with a brittle lacquer

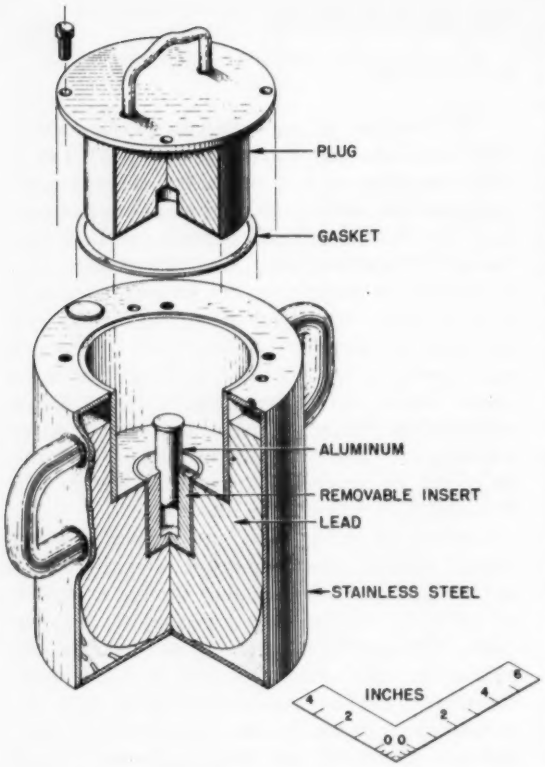


Fig. VIII-6 Returnable container for solid materials (lead shielding, 4 to 7½ in.).

to determine the areas of high stress and thereby indicate where some of the instrumentation was to be placed. The casks were dropped horizontally, on end, on an edge (corner), and on pistons, and each cask was dropped more than once.

As a result of the drops, many areas of the outer and inner shell took a slight but permanent set. Except for the actual impact area, the inner shell was subject to higher stresses than was a comparable position on the outer shell. The maximum permanent change of the inner diameter of the casks was 1 in. for both the small and large casks when dropped 20 to 30 ft. In no drop was the inner shell penetrated. A water-filled cask continued to hold water after a 15-ft horizontal drop.

Figure VIII-7 shows a 1.4-ton cask being dropped on a corner. The picture was made from selected frames taken by a high-speed motion-picture camera operating at about 3600 frames/sec. The sequence shows that the total impact, including bounces, occurs in about 0.5 sec. Following a 15-ft corner drop of a 1.4-ton cask coated with brittle lacquer, radial stresses were observed perpendicular to the cracking in the brittle lacquer material and were concentrated around the lead fill holes. After horizontal drops of a 1.4-ton cask from 15 and 28 ft, a small crack appeared in the weld where the outer shell and the end flange join. Cracks in the brittle lacquer, indicating high stress around the lead fill hole, were also seen. When a 1.4-ton cask was dropped 5 ft onto a 2-in.-diameter piston, the piston penetrated about 1.5 in. into the cask.

2. Fire Tests. In a 15-min fire test at 1320°F, made by Underwriters' Laboratories,[9] lead was ejected from the cask. In fire tests at ORNL, pressure from the vaporization of a small amount of water resulted in spraying of molten lead through pinholes in the top flange of the cask.[10] Therefore it is doubtful that unprotected lead-filled casks would meet fire requirements.[4]

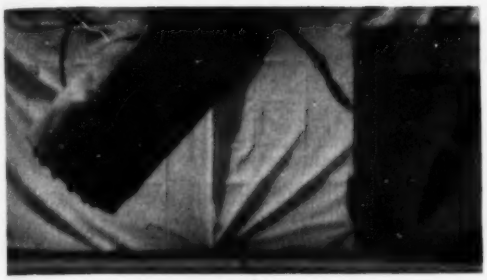
Wooden Fire and Impact Shields. A typical shield is 3⅝-in.-thick maple (Fig. VIII-8) with mitered corners of nailed construction. A mild-steel frame holds the shield intact under impact conditions. The frame is made of 1-in. steel straps and 1.5-in. steel angle iron for the small size, and up to 2.5-in. and 3-in. angle iron for the largest shield. The top is hinged for removal of the container.

The smallest shield tested weighs 120 lb and holds a lead-filled 60-lb steel container that has 1.5-in. shielding. The largest shield weighs 570 lb and holds a 2600-lb container with 8-in.-thick lead shielding.

Methods under consideration for protecting larger shields (larger than 1.5 tons) include shock-absorbing aluminum honeycomb.

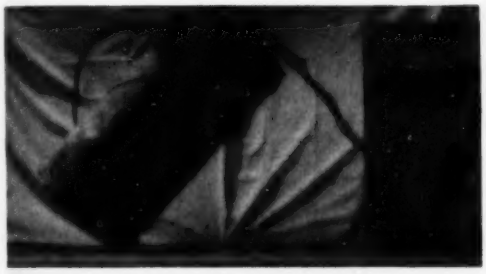
1. Structural Tests. The lead-filled steel cask models and the wooden fire and impact shields were dropped from heights up to 30 ft onto a pad consisting of a 12- by 12-ft piece of steel armor plate 4.5 in. thick, backed up by a 5-ft-thick slab of reinforced concrete. Below the pad was a 3-ft-diameter reinforced column reaching down approximately 7 ft to bedrock and then extending 3 ft into the bedrock.

The small wooden shield, 120 lb, which measures 14 by 14 by 17⅜ in., was dropped from heights of 15 and 30 ft onto a 6-in.-diameter piston. Very little damage was done to the shield,



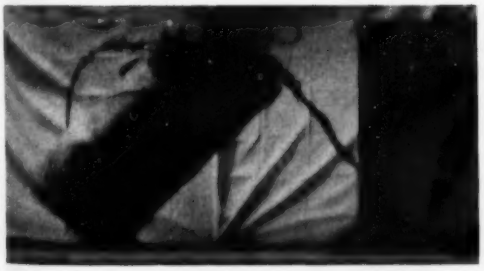
Prior to impact

Second impact—time: 0.102 sec



Initial impact—time: 0.000 sec

Second bounce—time: 0.196 sec



Peak deceleration—time: 0.0083 sec

Third impact—time: 0.391 sec

First bounce—time: 0.071 sec

Final position—time: 0.515 sec

Fig. VIII-7 Corner drop of a 1.4-ton model shipping cask (drop height, 15 ft).

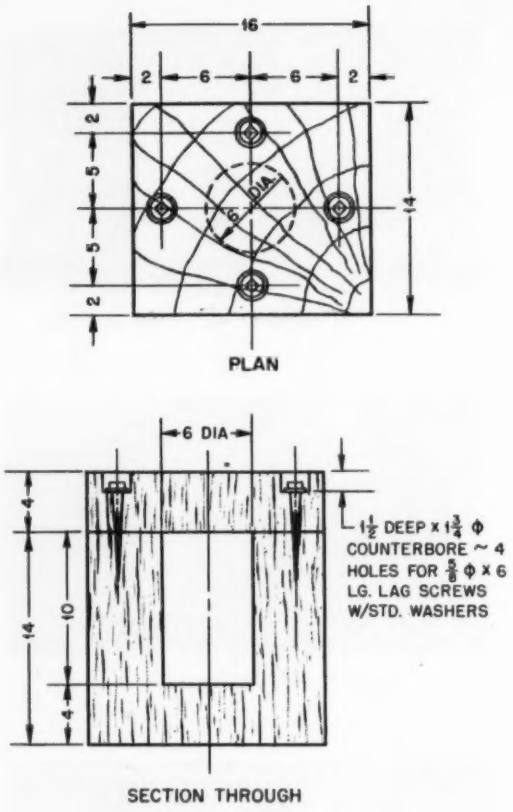


Fig. VIII-8 Wooden fire shield.

which was loaded with four lead bricks weighing a total of 112 lb to simulate the cask. There was some damage to the hinges of the top.

A second shield of the same size was dropped 30 ft onto a corner. There was no damage to the hinges, and damage to the rest of the shield was minimal.

The larger shields did not perform quite so well as the smaller ones, but they did meet current AEC (Ref. 4) and ICC (Ref. 5) regulations on structural requirements. The larger shield is designed for a 3.5-ft drop onto a 6-in.-diameter piston and a 30-ft free fall onto one corner. Testing on the larger shields is not yet complete.

Another design using a solid block of wood was tested in a 15-ft free fall, but, because of cracking and splitting of the wood, it was abandoned.

2. Fire-Resistance Tests. Fire-resistance tests were made on pine, hickory, maple, and ebony. Maple was selected because of its resistance to burning (a 2-in. penetration vs. 2.5 to 3 in. for pine in 1 hr), availability, strength, insulating properties, and cost. Ebony, of course, is too expensive and is a rather poor insulator. Hickory was not available in the thicknesses and widths needed and is no less expensive than maple. Pine was the best insulator, but it burned more rapidly and did not have the strength of maple.

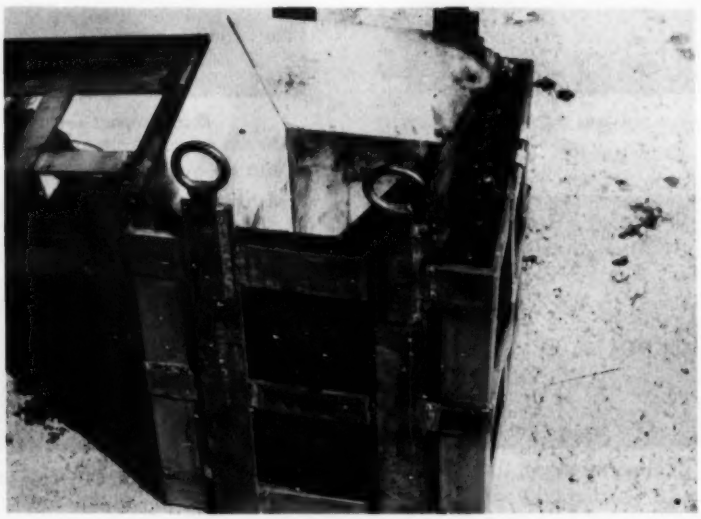


Fig. VIII-9 Fire and impact shield after a 1-hr fire test.

The shields were tested for 1 hr in the flame of a pressurized kerosene burner. The outside temperature was maintained at 900 ± 50°C for 1 hr to simulate the standard 1-hr fire test.

Pine shields burned 2.5 to 3 in. through. Maple burned about 2 in. Both appeared to be good insulators. After exposure to the fire for 1 hr, the maximum temperature in the pine shield rose to 30°C, and in the maple shield, to 60°C. Figure VIII-9 shows the maple shield after 1 hr of exposure to the fire.

The average cost of the shields will be approximately $250 each. The total cost represents 10% of the total cost of replacing all carriers now used and approximately 20% of the cost of modifying all the carriers now used in order to meet new safety criteria. A disadvantage is that freight costs will be increased from 20 to 200% on radioisotope shipments requiring this additional protection.

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Section

IX

Isotopes and Radiation Technology

Available Materials and Services

General Electric has established a new Irradiation Services and Products Section in its Atomic Power Equipment Department to meet the growing demand for radioisotopes and irradiation services. With headquarters at Vallecitos Atomic Laboratory (Pleasanton, Calif.), it will offer a complete range of nuclear irradiation services, including production of radioisotopes.

Union Carbide Nuclear Company has announced that interest in isotopes is sufficient to encourage the company "to become a major producer of isotopes and to provide consultation and services in radiation application problems." They are now supplying numerous routinely used isotopes and have indicated that they can prepare others in bulk quantities or as special materials. They also plan to quadruple their neutron activation analysis business.

In February 1963, bacon became the first food to be approved by the U. S. Food and Drug Administration for radiation sterilization. The bacon, which is irradiated in cans with 4.5 Mrads from a Co^{60} gamma source, will next be tested under arctic and tropical conditions. Some twenty other foods being studied for such processing include chicken, ham, potatoes, shrimp, and oranges.

In the meantime the U. S. Atomic Energy Commission (AEC) has negotiated with Stone & Webster Engineering Corporation to build the first seafood products irradiation pilot plant at Gloucester, Mass. The $600,000 plant, with a 300-kilocurie Co^{60} gamma source and a capacity of 1 ton/hr, will be used to demonstrate the feasibility of radiation pasteurization of seafoods. The AEC's 1964 budget includes $350,000 for a transportable Co^{60} irradiator for field studies with fruits and $200,000 for a grain-disinfestation pilot plant.

A Co^{60} irradiation plant has been opened in England by Johnson's Ethical Plastics Ltd. for sterilizing plastic disposable syringes. The source initially is a 50-kilocurie unit, but the design is for 250 kilocuries.

Export of radioisotopes will be simplified by recent amendments to AEC regulations. A general license—one for which no application to AEC is required—has been established to replace the specific license formerly required for the export of certain forms of moderate quantities of tritium and polonium. Use of these isotopes, which is widespread and considered safe in the atomic energy industry, will be subject to the radiation safety laws and regulations of the countries to which they are exported.

The sale price of heavy water has been cut from $28 to $24.50 per pound f.o.b. Savannah River Plant, Aiken, S. C. The base charge for leased heavy water has also been cut to $24.50 for leases that do not have a fixed base charge. Further price cuts will be considered if commitments from customers require operation of the heavy-water plant at increased capacity.

Oak Ridge National Laboratory has 99.99% Ne^{20} available for routine sales. In June 1963, concentrates of the mid-position isotopes, 20% Ne^{21} and 13% Ar^{38}, became available in very limited amounts.

The consultative committee for Eurisotop, the Euratom Information Bureau, held its first meeting in November 1962. A major topic of discussion was how to provide information on radioisotopes, not only to the general public, but to professional circles that may benefit from using them. Suggestions offered included traveling exhibits and television programs.

The AEC has executed agreements with Kentucky, Mississippi, California, New York, Texas, and Arkansas for transferring to the states some of its regulatory responsibility for licensing, rule-making, and enforcement in the uses of radioisotopes.

Oak Ridge National Laboratory's new edition of its Isotopes Catalog is now available. For the first time both stable and radioisotope offerings are included together. Worthy of note are the Commission's new low prices for bulk purchases of fission products: Sr^{90}, $0.75 per curie for >30 kilocuries; Cs^{137}, $0.50 per curie for >100 kilocuries; Ce^{144}, $1.00 per curie for >100 kilocuries. Cesium-137 at this price should be quite competitive with Co^{60} for many applications.

As of Aug. 12, 1963, the AEC (specifically ORNL and National Reactor Testing Station) terminated land burial services for licensees in favor of privately owned facilities such as Nuclear Engineering Co., Flemingsburg, Ky., and Beatty, Nev., and Nuclear Fuel Services in upper New York State.

In early June the AEC Regulatory Staff moved to Bethesda, Md. Their address is: Phillips Building, Corner St. Elmo and Norfolk Avenues.

The Second Annual Oak Ridge Radioisotopes Conference will be held Apr. 19–21, 1964, in Gatlinburg, Tenn. Cochairmen are R. T. Overman of Oak Ridge Institute of Nuclear Studies and A. F. Rupp of Oak Ridge National Laboratory.

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